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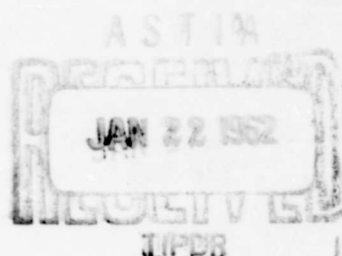
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**HIGH TEMPERATURE OXIDATION RESISTANT  
COATINGS FOR TANTALUM BASE ALLOYS**

DEAN D. LAWTHERS  
L. SAMA

SYLCOR DIVISION  
SYLVANIA ELECTRIC PRODUCTS, INC.

OCTOBER 1961



AERONAUTICAL SYSTEMS DIVISION

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*DEAN D. LAWTHERS  
L. SAMA*

*SYLCOR DIVISION  
SYLVANIA ELECTRIC PRODUCTS, INC.*

*OCTOBER 1961*

DIRECTORATE OF MATERIALS AND PROCESSES  
CONTRACT No. AF 33(616)-7462  
PROJECT No. 0(7-7351)  
TASK No. 73512

AERONAUTICAL SYSTEMS DIVISION  
AIR FORCE SYSTEMS COMMAND  
UNITED STATES AIR FORCE  
WRIGHT-PATTERSON AIR FORCE BASE, OHIO

## FOREWORD

This report was prepared by Sylcor Division, Sylvania Electric Products, Inc. under USAF Contract No. AF33(616)-7462. This contract was initiated under Project No. 0(7-7351), Task No. 73512. The work was administered under the direction of the Directorate of Materials and Processes, Deputy for Technology, Aeronautical Systems Division, with Mr. N. M. Geyer acting as project engineer.

This report covers work conducted from 1 June 1960 to 31 May 1961.

The program was carried out under the direction of Mr. L. Sama. The work was supervised by Mr. D. D. Lawthers. The aluminum coating phase was performed by Mr. G. T. Pepino, Jr., beryllium coating phase by Mr. D. Melone, hardness and flame oxidation testing by Mr. O. Haines, and metallography by Mr. H. Woods.

## ABSTRACT

Aluminide and beryllide coatings were investigated for pure tantalum, a commercial Ta-10 W alloy, and a ternary alloy under development. Coatings were applied by dipping, packing, cold spraying, and vapor deposition. Oxidation resistance was evaluated by furnace testing, resistance heating, and flame testing. Internal hardening and diffusion effects were also studied. A Sn-Al coating was developed, which has excellent oxidation resistance for 10 hours to at least 3000°F. Evaluation was successfully extended to arc-plasma tests which also showed the feasibility of protecting molybdenum and tungsten. A similar but minor effort investigation was performed with a columbium alloy which could be protected to 2500°F for at least 10 hours.

## PUBLICATION REVIEW

This report has been reviewed and is approved.

FOR THE COMMANDER:



I. PERLMUTTER

Chief, Physical Metallurgy Branch  
Metals and Ceramics Laboratory  
Directorate of Materials & Processes

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# HIGH TEMPERATURE OXIDATION RESISTANT COATINGS FOR TANTALUM BASE ALLOYS

## I. INTRODUCTION

The development of tantalum base alloys with attractive high temperature mechanical properties has been quite recent<sup>(1)</sup> and begins to make tantalum and its alloys appear promising for high temperature use, such as re-entry vehicles. Its high cost and density are somewhat offset by excellent ductility and fabricability. As with the other refractory metals tantalum has poor elevated temperature oxidation resistance<sup>(2)</sup>, especially in the range 2000°-3400°F where it may be expected to be useful. Therefore, the need for protective coatings is evident.

The program was intended to approach coating development from a fundamental viewpoint. Since tantalum alloys are contemplated for use in a variety of applications, coatings were not to be developed for any specific utilization, but rather to be sufficiently characterized to permit evaluation for the several potential applications. The capabilities of all experimental coatings were to be determined by subjecting coated tantalum specimens to at least the following evaluation tests:

1. Continuous isothermal exposure in air at temperatures from 2500° to 2800°F for a minimum of 10 hours.
2. Withstand at least 10 cycles during 10 hours in an air blast between test temperature (2500° to 2800°F) and ambient.
3. The coating had to withstand 2% deformation at temperature without causing oxidation failure.

The specific approach to be employed in developing oxidation resistant coatings was to base such coatings on intermetallic compounds of tantalum. Aluminides and beryllides were known to have this required oxidation resistance<sup>(3)</sup> and the intent was to meet the objectives by producing a two mil (0.002") coating of these compounds on the surface.

Another requirement was to use simple coating techniques since complex shapes or large pieces might be required in practical applications.

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## A. Materials

Coatings were evaluated on three tantalum base alloys: (1) pure tantalum; (2) a commercial Ta-10 W alloy; and (3) Ta-10 Hf-5 W alloy which was produced by Battelle Memorial Institute. Since some re-entry vehicles require thin sheets and to magnify diffusion effects, 0.010" thick sheet was used for the investigation. Alloys were coated in the steel grit-blasted, cold rolled, as-received condition. The corners and edges of the sample were not rounded. Several size samples were used for different testing techniques.

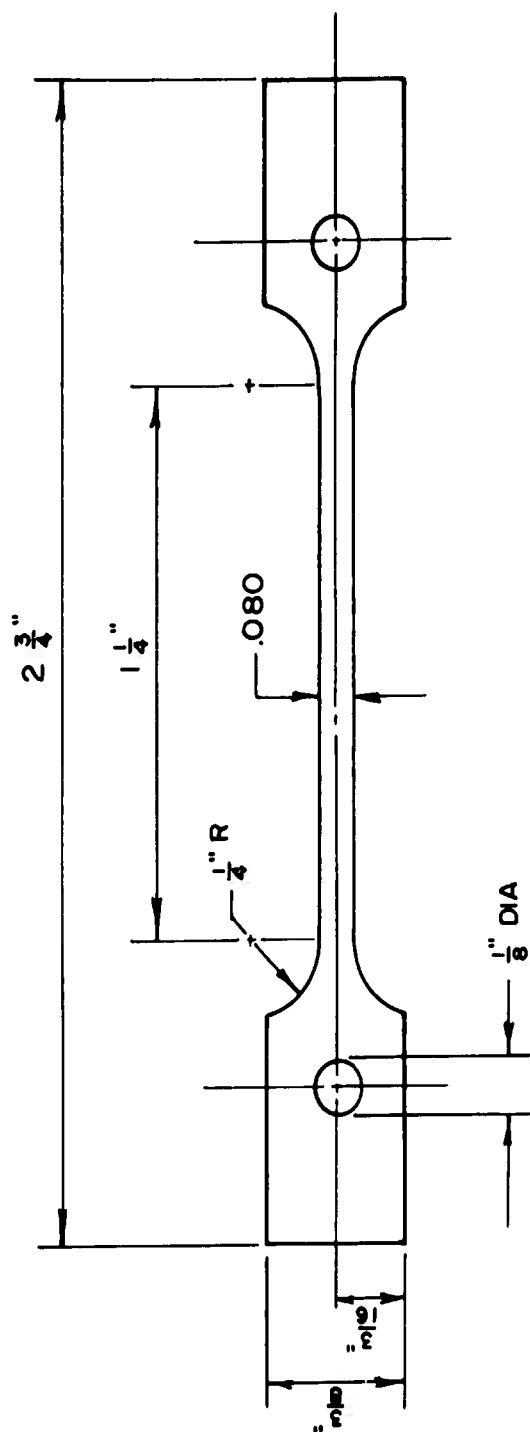
## B. Oxidation Procedure

To obtain oxidation results three different methods of heating were used. The most used technique was by electrically heating by direct resistance, sheet tensile type samples as shown in Figure 1. One of the four heating units is shown in Figure 2, and a schematic sketch in Figure 3. Current was fed to the sample through copper clamps. The lower clamp makes contact in a pool of mercury. Maximum current was 55 amperes for these units and the standard 2500°F test only requires about 100 watts. The temperature was measured with an optical pyrometer. Oxidation tests could be made either isothermally or by thermal cycling to room temperature with an air blast flowing, both under tensile loads. All compositions are given in weight percent in this report. The standard test sample was measured for width and thickness, the length between holes recorded to 0.0001" for strain purposes (length of hot zone was 0.40"), and the testing units programed for 10 hours oxidation time with 55 minutes between cycles (could be varied between 1 and 60 minutes).

A cycle consists of turning the power off and having the air blast hitting the sample for ten seconds before the current comes back on. This time could and was varied up to 30 seconds with no difference noted on oxidation behavior. Therefore, a ten second interval was arbitrarily selected. If the sample failed prematurely, a microswitch was actuated and shuts all power off. A normal 12 hour clock indicates the time of failure.

It is known that this type of heating has certain inherent disadvantages such as:

1. The inside is hotter than the surface,



Scale 2X

Fig. 1. Direct resistance tensile specimen.

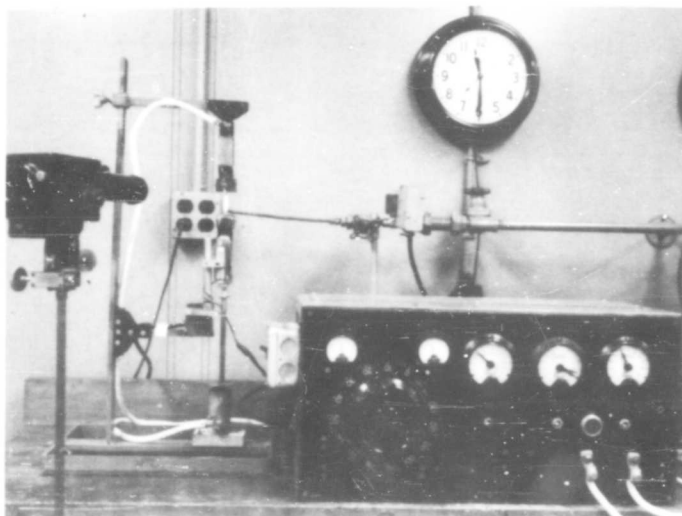


Fig. 2. Electrically heated oxidation test apparatus.

- |                |               |                      |
|----------------|---------------|----------------------|
| (A) HR. CLOCK  | (E) AMMETER   | (I) STRAIN INDICATOR |
| (B) MIN. CLOCK | (F) VARIAC    | (J) WEIGHT PAN       |
| (C) SEC. CLOCK | (G) SWITCHES  | (K) SAMPLE           |
| (D) VOLT METER | (H) AIR BLAST | (L) CLAMPS           |

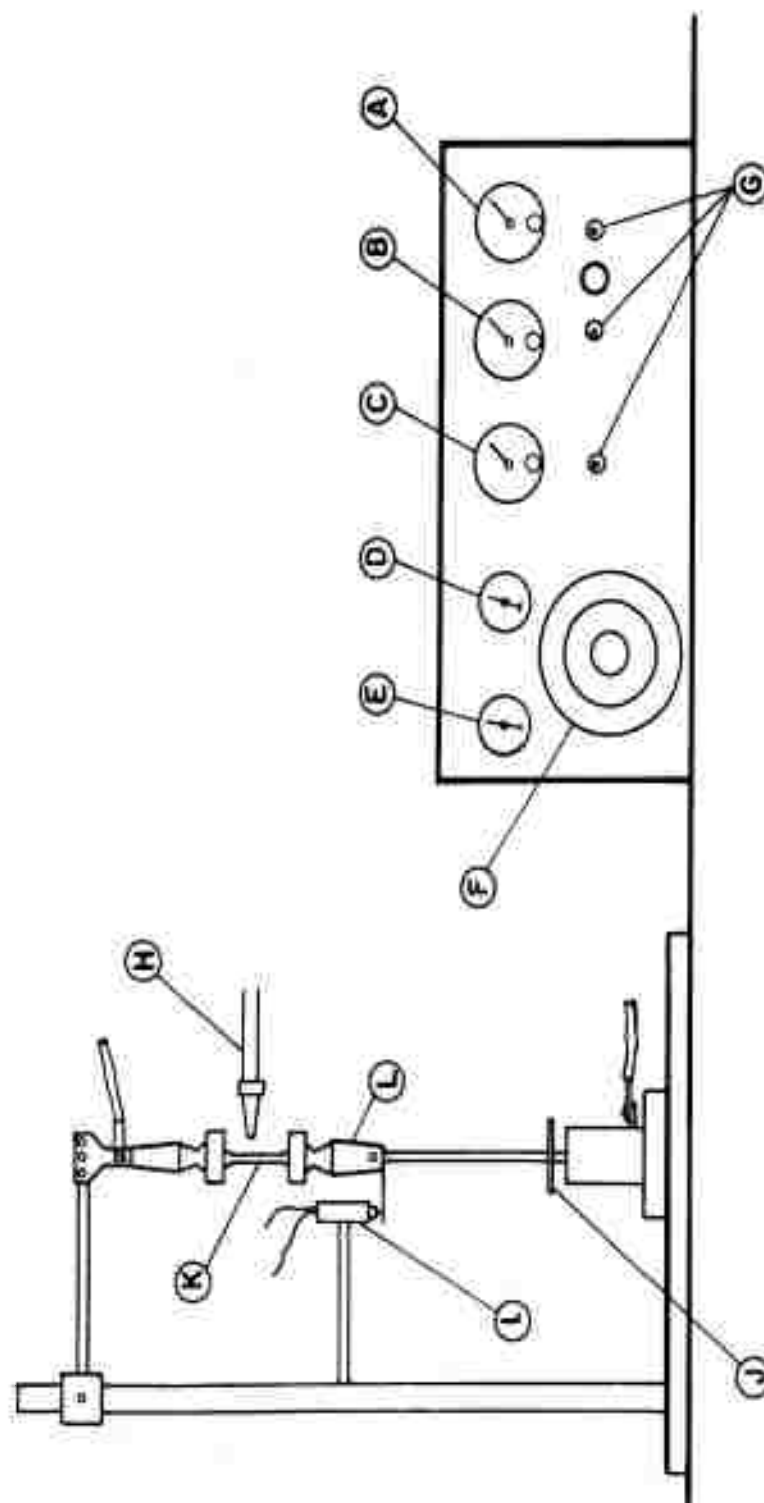


Fig. 3. Schematic of electrically heated oxidation test apparatus.

2. The emissivity of the surface can change as a function of time,
3. Any change that decreases current carrying area increases temperature and accelerates oxidation, and
4. Heating is from the inside out rather than from the outside as would be the case in actual use.

However, if good correlation could be found between furnace tests and direct resistance heating results, the advantage of economy, ease and flexibility of testing, visual observation of oxidation, and multiplicity of results from one test clearly outweigh the disadvantages. This type of oxidation test procedure gave a comparable evaluation of different coatings.

The second method of oxidation testing was to heat a 2" long x 1" wide x 0.010" thick piece of coated tantalum in an oxygen-gas flame. Again the temperature was measured by an optical pyrometer. The sample was cycled in an air blast. A sketch of the apparatus is given in Figure 4.

The third and most conventional manner of oxidation testing used was with coated 3/4" x 1/2" x 0.010" coupons in a furnace with slowly moving air. The samples could be removed periodically for thermal cycling and visual observation.

### C. Coating Techniques

Tantalum aluminide type coatings were produced by a number of different techniques as follows:

1. Vacuum or argon pack calorized using fine elemental powders,  $Al_2O_3$ , with or without a carrier such as  $NH_4Cl$ ;
2. Dipping in a molten aluminum alloy bath with or without a subsequent pack calorizing heat treatment;
3. Slurry painting with metal powders in a nitro-cellulose lacquer followed by a vacuum diffusion heat treatment;
4. Spraying the slurry followed by vacuum heat treatment; or
5. Combinations of the above.



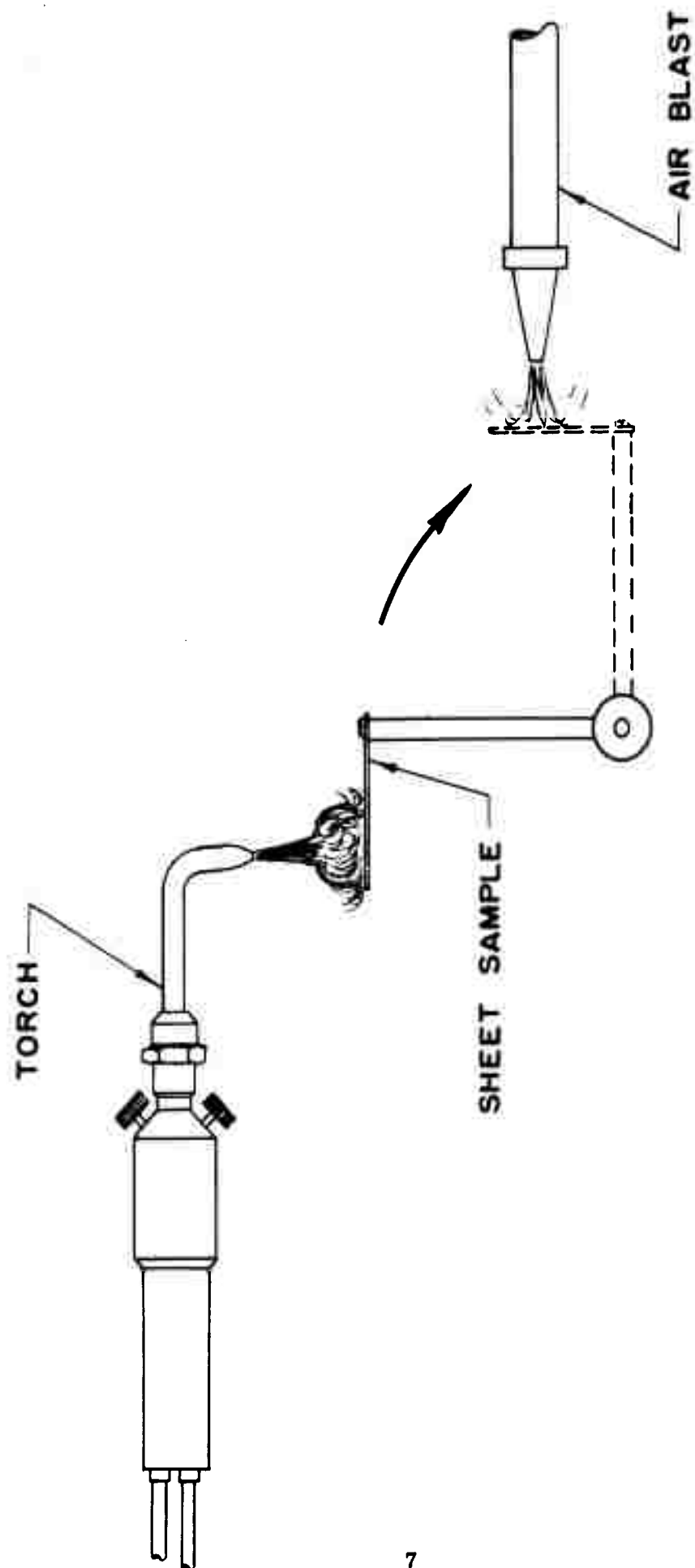


Fig. 4. Flame oxidation test rig.

It must be emphasized that coating compositions were varied mainly to suit a particular method of application and to obtain maximum oxidation characteristics. In all cases the end product desired was the tri-aluminide or aluminide plus excess tin coating.

## II. ALUMINUM COATED TANTALUM

The first step in the program was to investigate the formation of tantalum aluminide by hot dipping in molten aluminum and aluminum base alloys with and without subsequent reaction-diffusion treatment at elevated temperatures in inert atmospheres.

To determine the effect of dipping variables on coatings, the following program was established:

1. Dip in molten aluminum and aluminum alloys at 1700° and 2100°F for 1, 2, and 5 minutes followed by
2. A vacuum diffusion heat treatment, or
3. A pack calorizing heat treatment in Al or Al alloy plus  $\text{Al}_2\text{O}_3$  powders at 1900° and 2200°F for 1 and 4 hours.

All results on pure tantalum were obtained using annealed sheet. The test coupons were 0.5" x 0.75" x 0.010".

Samples were dipped in four baths: G3 pure aluminum, G4 Al-11 Si, G5 Al-10 Si-10 Cr, G6 Al-25 Si. The effect of test variables on coating thickness and solution of the tantalum core is given in Table 1. All measurements were made on metallographic samples. The original thickness of the tantalum sheet is 9.7 mils (0.0097"). The following statements can be made based on these results:

1. Using an aluminum bath, varying dipping time and temperature has no significant effect on the thickness of coating, which was one mil per side, but the tantalum core was dissolved away rapidly with increasing time and temperature.
2. A vacuum heat treatment cannot be used on dipped tantalum specimens because of the rapid vaporization of free aluminum before the intermetallic tantalum aluminide is formed.

TABLE 1

## THICKNESS OF ALUMINUM COATINGS ON TANTALUM SHEET

<u>Dipping</u>		<u>Thickness in mils</u>			
<u>Temp., °F</u>	<u>Time, Min.</u>	<u>Coating</u>	<u>Core</u>	<u>Coating</u>	<u>Total</u>
<u>Pure Aluminum Dip, As Dipped</u>					
1700	1	1.3	9.3		11.9
1700	2	1.3	9.1		11.7
1700	5	1.2	8.9		11.3
2100	1	1.7	8.6		12.0
2100	2	1.2	6.3		8.7
<u>Vacuum Diffused 1900°F 1 hour</u>					
1700	1	1.4	8.4	1.7	11.5
1700	2	1.4	8.4	1.6	11.4
1700	5	1.2	7.6	1.2	10.7
2100	1	1.1	7.6	1.2	9.9
2100	2	1.3	7.3	1.3	9.9
<u>Vacuum Diffused 1900°F 4 hours</u>					
1700	1	1.2	8.3	1.2	10.7
1700	2	1.2	8.3	1.2	10.7
1700	5	1.2	8.3	1.2	10.7
2100	1	1.0	7.8	1.2	9.8
2100	2	0.62	7.1	1.2	8.3
<u>Vacuum Diffused 2200°F 1 hour</u>					
1700	1	1.1	8.4		10.6
1700	2	1.1	8.4		10.6
1700	5	1.1	8.4		10.6
2100	1	0.99	7.7		9.7
2100	2	0.76	6.4		7.6
<u>Vacuum Diffused 2200°F 4 hours</u>					
1700	1	1.0	8.2	1.0	10.3
1700	2	1.0	8.4	0.74	10.1
1700	5	0.74	8.2	0.49	9.4
2100	1	0.49	7.9	0.44	8.8
2100	2	0.45	6.9	0.45	7.8

TABLE 1 (Cont'd.)

## THICKNESS OF ALUMINUM COATINGS ON TANTALUM SHEET

<u>Dipping</u>		<u>Thickness in mils</u>			
<u>Temp., °F</u>	<u>Time, Min.</u>	<u>Coating</u>	<u>Core</u>	<u>Coating</u>	<u>Total</u>
<u>Pack Calorized 1900°F 1 hour</u>					
1700	1	1.9	8.1	1.9	11.9
1700	2	2.1	7.9	1.7	11.7
1700	5	2.1	7.8	1.7	11.6
2100	1	1.9	7.4	2.0	11.3
2100	2	2.5	6.2	2.2	10.9
<u>Pack Calorized 1900°F 4 hours</u>					
1700	1	2.1	7.7	2.0	11.8
1700	2	1.4	7.8	1.7	10.9
1700	5	1.2	7.9	1.2	10.3
2100	1	1.2	7.4	1.6	10.2
2100	2	1.3	6.8	1.3	9.4
<u>Pack Calorized 2200°F 1 hour</u>					
1700	1	1.6	7.8		
1700	2	1.7	7.7		
1700	5		7.7		
2100	1		7.2		
2100	2		5.8		
<u>Pack Calorized 2200°F 4 hours</u>					
1700	1	1.7	7.4	2.0	11.1
1700	2	2.0	7.3	1.9	11.2
1700	5	1.4	7.7	1.4	10.5
2100	1	1.4	6.9	1.6	9.9
2100	2	1.7	6.2	1.7	9.6
<u>Al-11 Si Dip Vacuum Diffused 1900°F 1 hour</u>					
1700	1	1.7	8.1	1.4	11.2
1700	2	0.74	8.3	0.74	9.7
1700	5	0.37	8.3	0.37	9.0
2000	0.5	1.4	8.1	1.2	10.7
2000	1	2.1	7.2	2.1	11.4

TABLE 1 (Cont'd.)

## THICKNESS OF ALUMINUM COATINGS ON TANTALUM SHEET

<u>Dipping</u>		<u>Thickness in mils</u>			
<u>Temp., °F</u>	<u>Time, Min.</u>	<u>Coating</u>	<u>Core</u>	<u>Coating</u>	<u>Total</u>
<u>Vacuum Diffused 1900°F 4 hours</u>					
1700	1	1.3	8.3	1.0	10.6
1700	2	0.49	8.2	0.49	10.1
1700	5	0.4	8.7	0.4	9.5
2000	0.5	1.6	7.7	1.4	10.1
2000	1	0.3	6.9	2.7	12.6
<u>Vacuum Diffused 2200°F 1 hour</u>					
1700	1	1.0	8.1	1.0	10.1
1700	2	1.0	8.2	1.0	10.2
1700	5	1.1	8.4	1.0	10.5
2000	0.5	1.4	8.2	1.2	10.8
2000	1	2.6	7.1	2.5	12.2
<u>Vacuum Diffused 2200°F 4 hours</u>					
1700	1	2.0	7.2	2.0	11.2
1700	2	1.2	8.1	2.1	11.4
1700	5	0.74	8.3	0.74	9.7
2000	0.5	1.2	7.8	1.2	10.2
2000	1		9.1		
<u>Pack Calorized 1900°F 1 hour</u>					
1700	1	1.9	7.6	1.9	11.4
1700	2	2.2	7.4	2.1	11.7
1700	5	2.2	7.4	2.0	11.6
2000	0.5	2.0	7.8	1.9	11.7
2000	1	2.0	7.4	1.9	11.3
<u>Pack Calorized 1900°F 4 hours</u>					
1700	1	2.5	7.4	2.0	11.9
1700	2	2.2	7.7	2.0	11.9
1700	5	1.9	7.7	1.4	11.0
2000	0.5	1.4	7.8	1.7	10.9
2000	1	2.7	6.8	3.0	12.5

TABLE 1 (Cont'd.)

## THICKNESS OF ALUMINUM COATINGS ON TANTALUM SHEET

<u>Dipping</u>		<u>Thickness in mils</u>			
<u>Temp., °F</u>	<u>Time, Min.</u>	<u>Coating</u>	<u>Core</u>	<u>Coating</u>	<u>Total</u>
<u>Pack Calorized 2200°F 1 hour</u>					
1700	1	2.9	7.4	3.0	13.3
1700	2	5.2	5.0	5.1	15.3
1700	5	2.2	7.4	2.2	11.8
2000	0.5	1.4	7.6	1.2	10.2
2000	1	1.9	7.2	2.0	11.1
<u>Pack Calorized 2200°F 4 hours</u>					
1700	1	2.5	7.2	2.4	12.1
1700	2	2.9	6.7	2.7	12.3
1700	5	2.5	6.8	2.5	11.8
2000	0.5	2.5	7.0	2.4	11.9
2000	1	2.6	6.5	2.5	11.6
<u>Al-10 Si-10 Cr Dip, Pack Calorized 1900°F 1 hour</u>					
1700	1	2.0	7.4	1.9	11.3
1700	2	2.6	7.4	0.49	10.5
2000	0.5	1.7	7.4	1.4	10.5
2000	1	3.7	5.7	4.0	13.4
<u>Al-25 Si Dip, Pack Calorized 1900°F 1 hour</u>					
1700	1	2.2	6.8	3.2	12.0
1700	2	1.6	6.8	2.9	11.3
2000	0.5	2.0	7.4	1.7	11.1
2000	1	3.3	6.2	3.8	13.3

3. Therefore, a pack calorizing heat treatment in argon, for example, must be used in order to form tantalum aluminide ( $\text{TaAl}_3$ ) on the surface. Pack calorizing consists of placing the dipped sample in a powder mixture of 10 Al, balance  $\text{Al}_2\text{O}_3$ , heating to some temperature, say  $1900^\circ\text{F}$ , for 1 hour in an argon atmosphere to react and diffuse free aluminum to form  $\text{TaAl}_3$  type compounds.
4. Dipping in Al-11 Si results in a better coating than Al. The reaction rate was much faster than with Al and the growth rate of the aluminide coating is time and temperature dependent.
5. A 2 mil coating seems to be needed for oxidation resistance under the test conditions. This will be discussed further.

Photomicrographs and hardness values for aluminum coated tantalum samples dipped for 1 minute at  $1700^\circ\text{F}$  with various diffusion treatments are given in Figures 5 to 8. Ideally under equilibrium conditions there should be three intermetallic compounds in the Ta-Al system,  $\text{TaAl}_3$  on the outer surface, then  $\text{Ta}_2\text{Al}$  and  $\text{Ta}_3\text{Al}$  adjacent to the tantalum core. The latter two compounds cannot be easily separated in the microstructure. Microstructures using other dipping baths of aluminum alloys are very similar. Figure 5 shows the unreacted aluminum on the tantalum surface. There is an unresolved zone of  $\text{TaAl}_3$  between the tantalum and the aluminum coating. Figures 6 to 8 reveal the growth of Ta-Al compounds as a function of time, temperature, and type of heat treatment. Parts of Figures 7 and 8 were taken under polarized light to show how optically active  $\text{TaAl}_3$  is. It became known from subsequent work that  $\text{TaAl}_3$  must be present initially to confer elevated temperature oxidation resistance.

A total of over 50 different aluminum compositions and/or application methods were investigated in this study. These are listed in Table 2. One general conclusion can be made at this point which will be proved by experimental evidence: only those aluminum compositions that contain tin will form the most satisfactory protective oxidation resistant coatings on tantalum. A double coating-diffusion process is also necessary for maximum protection.

Of all of the tin-free aluminum compositions probably only one could have practical significance. That would be the 90 Al-5 Ti-5

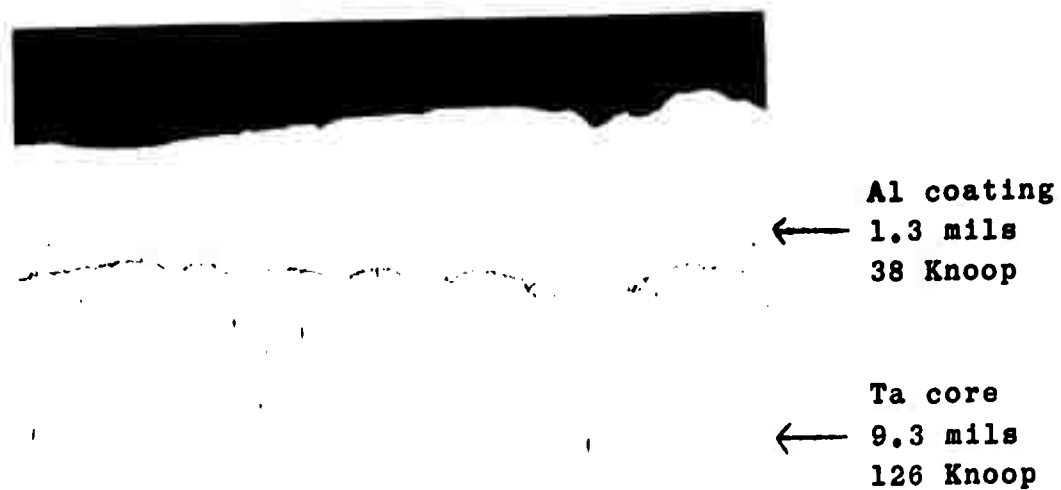


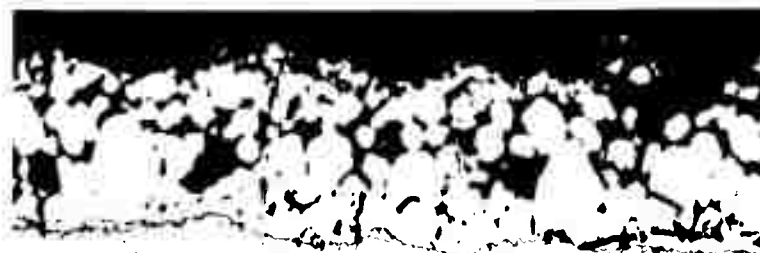
Plate No. 23383

G3171

500X

Fig. 5. Aluminum coated tantalum sheet  
as dipped 1 minute at 1700°F.





Al coating  
1.7 mils  
535 Knoop  
717 Knoop

Ta core  
8.4 mils

Plate No. 23401      G3171VD191      500X  
Vacuum diffused 1900°F 1 hour



Al coating  
1.2 mils  
480 Knoop

Ta core  
8.3 mils

Plate No. 23382      G3171VD194      500X  
Vacuum diffused 1900°F 4 hours

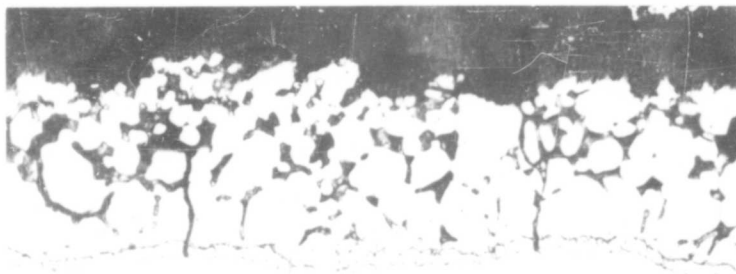


Al coating  
1 mil  
719 Knoop

Ta core  
8.2 mils  
114 Knoop

Plate No. 23384      G3171VD224      500X  
Vacuum diffused 2200°F 4 hours

Fig. 6. Aluminum coated tantalum sheet dipped  
for 1 minute at 1700°F, vacuum diffused.

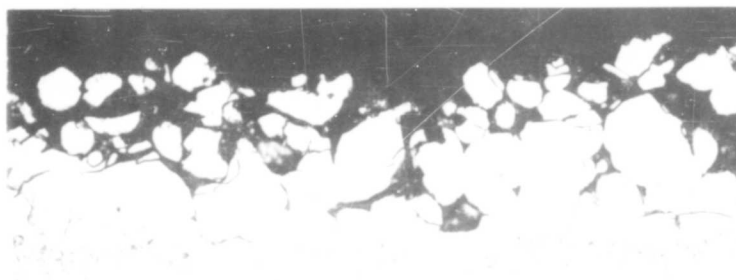


Al coating  
1.9 mils  
465 Knoop

← 678 Knoop

Ta core  
8.1 mils  
118 Knoop

Plate No. 23397      G3171PC191      500X  
Pack calorized 1900°F 1 hour



Al coating  
2 mils  
455 Knoop

← 697 Knoop

Ta core  
7.7 mils

Plate No. 23381      G3171PC194      500X  
Pack calorized 1900°F 4 hours

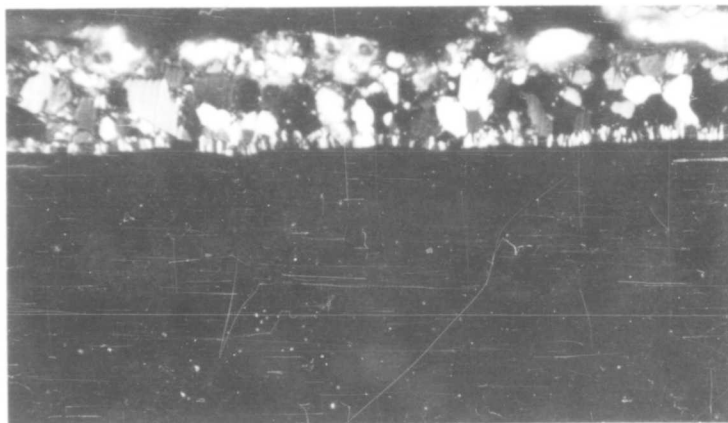


Plate No. 23387      G3171PC194      500X  
Pack calorized 1900°F 4 hours  
polarized light

Fig. 7. Aluminum coated tantalum sheet dipped  
1 minute at 1700°F, pack calorized



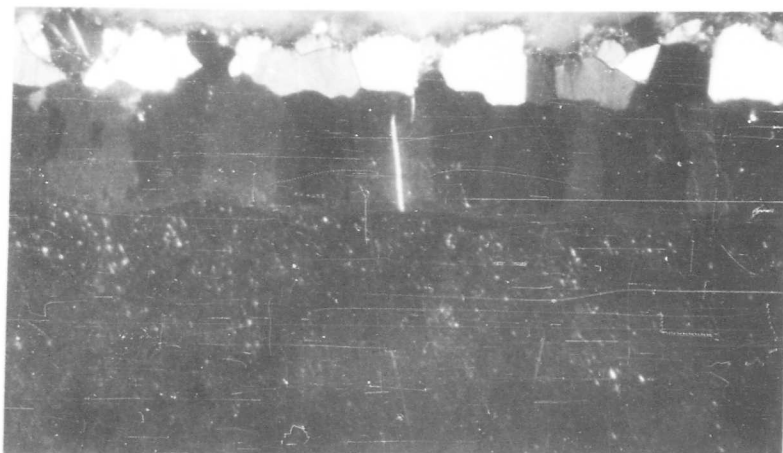
←  $TaAl_3$  469 Knoop  
 ←  $Ta_2Al$  916 Knoop  
 ← Ta edge  
 119 Knoop  
 ← Ta center  
 108 Knoop

Plate No. 23677

500X

Sample G3171PC221

Pack calorized 2200°F 1 hour, polarized light.



←  $TaAl_3$  493 Knoop  
 ←  $Ta_2Al$  1102 Knoop  
 ← Ta edge  
 124 Knoop  
 ← Ta center  
 115 Knoop

Plate No. 23676

500X

Sample G3171PC224

Pack calorized 2200°F 4 hours, polarized light.

Fig. 8. Aluminum coated tantalum sheet dipped  
 for 1 minute at 1700°F, pack calorized.

TABLE 2  
COATING COMPOSITIONS

<u>Code</u> <u>Identifi-</u> <u>cation</u>	<u>Composition, %</u>	<u>Method of</u> <u>Application</u>
G3	Pure aluminum	Molten Dip
G4	Al-11 Si	Molten Dip
G5	Al-10 Si-10 Cr	Molten Dip
G6	Al-25 Si	Molten Dip
G7	84.5 Al 10.5 Si 5 Y	Molten Dip
G8	71 Al 15 Cr 9 Si 5 Sn	Molten Dip
G9	90 Sn 10 (Al-10 Si)	Molten Dip
G10	71 Al 15 Cr 9 Si 5 Y	Molten Dip
G11	66 Al 15 Cr 9 Si 5 Y 5 Sn	Molten Dip
G12	90 Cu 10 (Al-10 Si)	Molten Dip
G13	72 Cu 22 Sb 6 (Al-10 Si)	Molten Dip
G14	90 Al 5 Cr 5 Ti	Molten Dip
G15	95 Al 5 Cr	Molten Dip
G16	95 Al 5 Ti	Molten Dip
G17	95 (Al-10 Si) 5 Ti	Molten Dip
G18	90 (Al-10 Si) 5 Cr 5 Ti	Molten Dip
G19	80 Al 13 Cr 4 Sn 3 Ti	Molten Dip
G20	58.4 Cu 34.4 Sb 7.2 (90 Al-5 Ti-5 Cr)	Molten Dip
G21	90 Sn 10 Al	Molten Dip
G22	90 Al 5 Cr 5 TiH <sub>2</sub>	Paint
G23	85 Al 10 Ti H <sub>2</sub> 5 Cr	Paint
G24	83 Al 10 Ti H <sub>2</sub> 5 Cr 2 Ta	Paint
G25	80 Al 10 Ti H <sub>2</sub> 5 Cr 5 Sn	Paint
G26	90 Sn 10 (90 Al-5 Ti H <sub>2</sub> 5 Cr)	Paint
G27	85 Al 10 Cr 5 Ti H <sub>2</sub>	Paint
G28	85 Al 5 Cr 5 Ti H <sub>2</sub> 5 Sn	Paint
G29	89 Al 5 Sn 3 Ti H <sub>2</sub> 3 Cr	Paint
G30	85 Al 5 Sn 5 Cr 5 Ti	Molten Dip
G31	85 Al 5 Sn 5 Cr 5 Ti H <sub>2</sub>	Paint
G32	94 Al 3 Ti 3 Cr	Molten Dip
G33	85 Al 5 Ti 5 Cr 5 Y	Molten Dip
G34	50 Al 50 Sn	Paint or Spray
G35	10 Al 90 Sn	Paint
G36	50 (90 Al-5 TiH <sub>2</sub> -5 Cr) 50 Sn	Paint or Spray
G37	85 Al 5 Ti 5 Cr 5 La	Molten Dip
G38	80 (90 Al-5 TiH <sub>2</sub> -5 Cr) 20 Al <sub>2</sub> O <sub>3</sub>	Vacuum Pack Calorized
G39	40 Al 58 Al <sub>2</sub> O <sub>3</sub> 2 NH <sub>4</sub> Cl	Argon Pack Calorized

TABLE 2 (Cont'd.)

## COATING COMPOSITIONS

<u>Code Identi- fication</u>	<u>Composition, %</u>	<u>Method of Application</u>
G40	25 Al 75 Sn	Paint or Spray
G41	75 Al 25 Sn	Paint or Spray
G42	50 Al 50 Ag	Paint
G43	33-1/3 Al 33-1/3 Sn 33-1/3 Cb	Paint
G44	33-1/3 (Al-11 Si) 33-1/3 Sn 33-1/3 Cb	Paint
G45	100 Cb	Spray
G46	65 Al 20 Cr 10 Sn 5 Si	Paint
G47	75 (90 Al-5 TiH <sub>2</sub> -5 Cr) 25 Sn	Paint or Spray
G48	40 (90 Al-5 TiH <sub>2</sub> -5 Cr) 58 Al <sub>2</sub> O <sub>3</sub> 2 NH <sub>4</sub> Cl	Argon Pack
G49	50 (90 Al-5 TiH <sub>2</sub> -5 Cr) 50 Cu	Spray
G50	5 Al 95 Sn	Molten Dip
G51	80 Al 5 Ti 5 Cr 10 Sn	Molten Dip
G52	50 Al 50 Sn	Molten Dip
G53	25 Al 75 Sn	Molten Dip
G54	50 (Al-11 Si) 50 Sn	Paint

Cr composition (G14) applied by hot dipping at 1900°F for 30 seconds followed by an argon diffusion treatment of 1 hour at 1900°F in a powder mixture of 10 Al and 90 Al<sub>2</sub>O<sub>3</sub>. But, as with all other tin-free aluminum compositions investigated in this program, the G14 coating has poor low temperature oxidation resistance in the range from 1100° to 1800°F and has poor thermal cycling oxidation resistance between test temperature (2500° to 2800°F) and ambient.

#### A. Compound Studies

Therefore, it was decided to obtain base line data on the oxidation characteristics of various compositions in the Ta-Al system. Arc-melted samples of TaAl<sub>3</sub>, "TaAl", Ta<sub>2</sub>Al, Ta<sub>3</sub>Al, and Ta<sub>0.9</sub>Ti<sub>0.1</sub>(Al<sub>0.9</sub>Si<sub>0.1</sub>)<sub>3</sub> were prepared and oxidized at 1100°, 1400°, 1800°, 2300°, and 2500°F.

The following conclusions can be made:

1. The oxidation results vary with location of coupon taken from the arc-melted sample.
2. None of these materials is single-phase as melted, probably caused by aluminum loss during arc-melting. It is known that there is no intermetallic compound of the composition TaAl<sup>(4)</sup>.
3. Ta<sub>2</sub>Al and Ta<sub>3</sub>Al are not oxidation resistant at any temperature tested.
4. TaAl<sub>3</sub> and the particular alloyed compound tested are not oxidation resistant at 1400°F but do have satisfactory potential at 2500°F.

This information helps to explain the low temperature anomaly and gives an indication of what to expect with tantalum aluminide type coatings. The 1400°F rapid oxidation was expected from later work on the columbium-aluminum system. But it was also assumed that tantalum trialuminide (TaAl<sub>3</sub>) would be more stable than columbium trialuminide (CbAl<sub>3</sub>). CbAl<sub>3</sub> was still present after 10 hours of air oxidation at 2600°F with aluminum coated columbium materials. The same was not true with aluminum coated tantalum materials. In fact times of less than one hour at temperatures as low as 2000°F in any atmosphere, under certain conditions, are enough to convert the TaAl<sub>3</sub> coating phase on tantalum to Ta<sub>2</sub>Al and Ta<sub>3</sub>Al. The effect

is shown in Figures 7 and 8 comparing a 4-hour diffusion heat treatment at 1900° and 2200°F. At the lower temperature the coating consists of almost entirely  $TaAl_3$  but at 2200°F little is left.

The  $TaAl_3$  phase must be present to form an oxidation resistant coating. This phase is reduced by the oxygen in the air at test temperature to form the protective  $Al_2O_3$  outer layer and  $Ta_2Al$  and  $Ta_3Al$ . Without going into detail at this point of the discussion the foregoing should give an idea of the problems to be met and solved to produce aluminum type coatings on tantalum that will protect it for at least 10 hours with a minimum of 10 thermal cycles under mechanical stress at test temperatures up to 2800°F.

### B. Oxidation Results

Since hundreds of oxidation tests have been run the individual results of such tests would fill dozens of tables and might prove tedious for the reader to cover. It appears preferable to give a concise appraisal and comparison of the general trends in oxidation behavior for the numerous combinations of coatings listed in Table 2.

Hot dipping in small aluminum alloy baths was used initially for screening purposes because of the ease of changing compositions, temperature, and time of coating. Later other application techniques were used. Post coating diffusion heat treatments were also investigated.

It was found that a 1700°F dipping temperature was too low to give the required oxidation protection at 2500°F to tantalum when dipped into (a) pure aluminum (G3), (b) Al-11 Si (G4), (c) Al-11 Si-5 Y (G7), or (d) Al-25 Si (G6). Tantalum samples dipped into pure aluminum (G3) at 2100°F without a diffusion heat treatment would withstand 4 hours at 2500°F isothermally. Failure occurred at longer times. Al-11 Si-5 Y coated samples (G7) dipped at 2000°F for 1 minute followed by a pack carburizing diffusion heat treatment of 1 hour at 1900°F increased oxidation protection to at least 8 hours at 2500°F but failure always occurred after 1 cycle to room temperature and reheating to 2500°F. The addition of 25 Si to the aluminum bath (G6) resulted in a non-adherent scale when oxidized at 2500°F. The Al-10 Si-10 Cr composition (G5) gave little added improvement in oxidation protection. The Al-15 Cr-9 Si-5 Sn composition (G8) would give 10 hours protection at 2500° and 2600°F with a few thermal cycles. All of these coating compositions would fail in a few hours if oxidized at 1100°, 1400°, or 1800°F.

The mechanism for this anomalous behavior is not known. However, it is postulated that the major factor is an expansion mismatch between the original  $TaAl_3$  intermetallic and the alloy substrate, the former having the higher coefficient. On cooling from the formation temperature to ambient temperatures the brittle surface compound is put in tension and cracks. Upon reheating to temperatures below the formation temperature the cracks do not close up and the substrate with its poor oxidation resistance is severely attacked. If the material is heated above its formation temperature the cracks close up and the surface is put in compression. There is also the possibility that diffusion of aluminum is not rapid enough at lower temperatures for the protective oxide film to repair itself or that a nonprotective porous film is formed at lower temperatures.

The beneficial effect of tin additions to aluminum alloy compositions as applied to tantalum was first shown with the Al-15 Cr-9 Si-5 Sn composition (G8). Yttrium was added to this composition with and without tin (G10 and G11) but no improvement in oxidation behavior was obtained. The 90 Sn-10 (Al-11 Si) composition (G9) caused a major breakthrough in oxidation protection. It will be discussed in a later section. Substituting copper or copper and antimony (G12 and G13) for tin as the diluent did not show too much promise.

The G14 composition, 90 Al-5 Ti-5 Cr, was the best tin free aluminum alloy found for protecting tantalum and the Ta-10 W alloy in the temperature range of 2500° to 2800°F. Over 150 samples of tantalum materials were oxidation tested in a furnace or by resistance heating. Samples electrically heated could take 10 hours and 10 cycles at 2500°, 2600°, and 2700°F under a stress to produce from 2 to 14% elongation without oxidation failure. Samples could also withstand 10 hours at 2800°F but cycling greatly reduced its life. Although furnace tested samples would last 10 hours at 2500° and 2600°F with cycles, behavior was more erratic. This was probably caused by sintering of the  $Al_2O_3$  in the coating to the recrystallized alumina boats with resultant rupturing of this oxide coating when handled for inspection. The time and temperature of use was increased by supporting the samples with platinum wires. In fact one sample lasted 308 hours at 2500°F isothermally without failure. This also points up the increased life without thermal cycling. This same coating applied by painting on the metal powders followed by a vacuum diffusion heat treatment (G22) had similar oxidation properties. Coating thickness was 1.5 mils.



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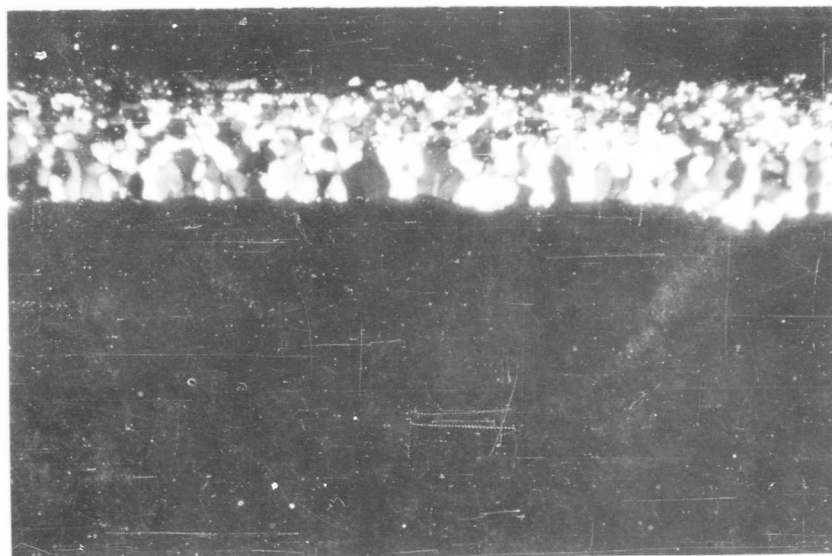
Effort was spent in varying the basic composition of G14 to try to improve properties. The addition of 5% tin seemed to increase the strength by reducing the alloy core loss in thickness as a function of time and temperature. In general, no improvement was found in any other composition change from 90 Al-5 Ti-5 Cr. This statement rules out coating composition and/or method of application G15, 16, 17, 18, 19, 20, 23, 24, 25, 27, 28, 29, 30, 31, 32, 33, 37, and 51. In any case the G14 composition also suffers from reduced low temperature oxidation resistance. This is not meant to rule out any aluminide composition as having poor high temperature oxidation behavior because G14 does have very good properties. But once again it must be reaffirmed that the Sn-Al coatings are much easier to apply and use for practical applications without any peculiar anomalies, at least on certain tantalum alloys.

Three coatings were applied by pack calorizing techniques. The samples were heated in a powder mixture for four hours at 1900°F (G38, 39, and 48). Samples would last for 16 hours at 2500° and 2600°F without failure but one thermal cycle was enough to cause failure. A second coating overlay of Sn-Al improves cyclic behavior but not enough to warrant additional work at this time.

Because of the rapid dissolution of  $TaAl_3$  and the known stability of  $CbAl_3$ , attempts were made to apply Cb-Al-Sn coatings directly to tantalum (G43 and G44) or to put a Cb layer on the Ta (G45) and coat. The first technique did not work and the second idea might work with more effort. This would involve finding more successful coatings for columbium which is not believed to be as practical as coating tantalum at this time.

### C. Metallographic Results on Al Coatings

Metallography has been a valuable tool in the present coating work. Microstructures together with Knoop hardness values (25 gram load) graphically show movements of compound interfaces, solution of tantalum, oxide growth, and serve to identify intermetallic compounds. The effect of oxidizing time and temperature on G14 coated pure tantalum are shown in Figure 9. After only one hour of oxidation at 2500°F, the  $TaAl_3$  has disappeared. Hereafter, the major portion is  $Ta_2Al$  which contains considerable porosity with the  $Ta_3Al$  thickness increasing with time. The dense  $Al_2O_3$  film can be seen and it can be noted that the oxide layer is thin and grows very slowly. The sintering of the porous  $Ta_2Al$  at higher temperatures and longer times is also noteworthy. Although tantalum is being dissolved, the rate is relatively slow. Oxidation times of



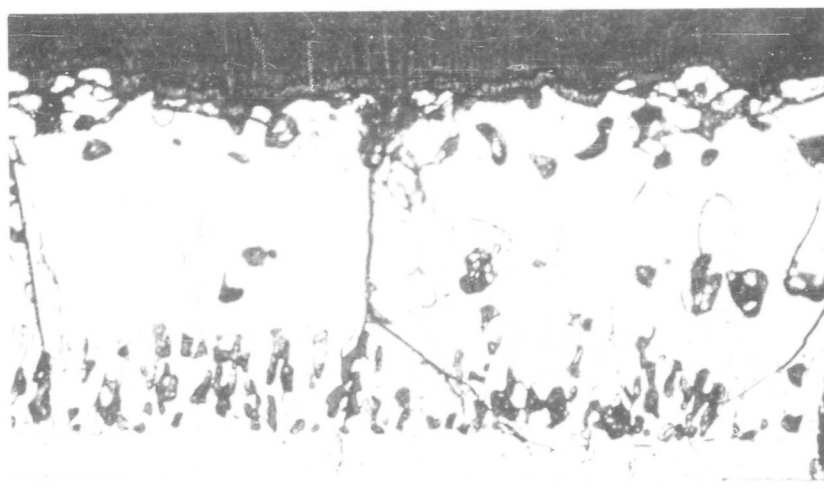
Knoop    Mils

←TaAl<sub>3</sub>    559    3.2

←Ta<sub>Edge</sub>    130  
Center    129    9.9

TOTAL    16.1

Plate No. 23667    Sample G14B19-1/2PC191    250X  
Polarized light diffused 1 hour 1900°F



←Ta<sub>2</sub>Al    1120    4.2

←Ta<sub>3</sub>Al    1160

←Ta<sub>Edge</sub>    141  
Center    132    6.9

TOTAL    16.0

Plate No. 23680    Sample G14B19-1/2PC191-44    500X  
Bright field oxidized 1 hour 2500°F

Fig. 9a.    Effect of oxidation time and temperature on 90 Al-5 Ti-5 Cr coated tantalum.



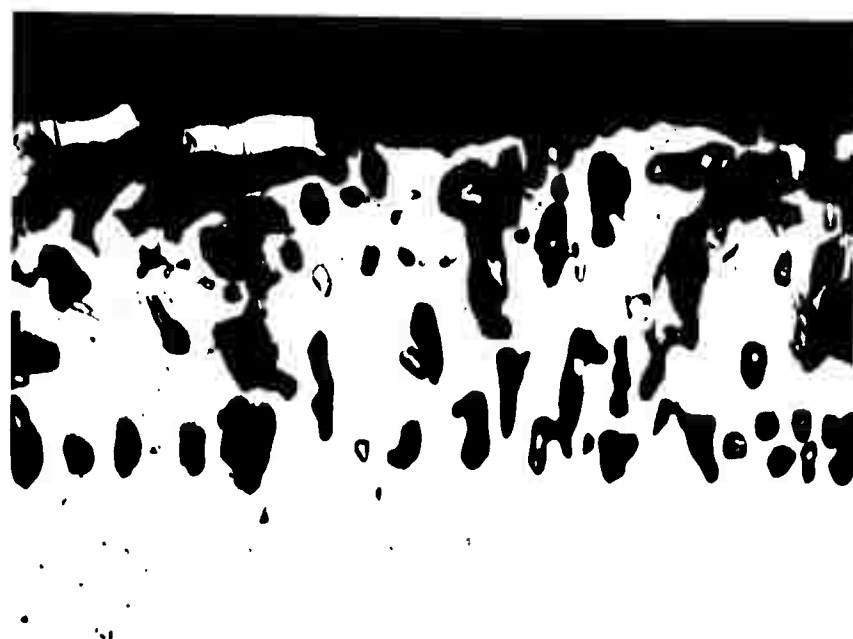
	<u>Knoop</u>	<u>Mils</u>
← Ta <sub>2</sub> Al	1184	5.5
← Ta <sub>3</sub> Al	1280	
← Ta	Edge 157 Center 148	6.1
TOTAL		16.5

Plate No. 23679

500X

Sample G14B19-1/2PC191-52

Fig. 9b. Oxidized 10 hours at 2500°F.



	<u>Knoop</u>	<u>Mils</u>
← $\text{Al}_2\text{O}_3$	4630	
← $\text{Ta}_2\text{Al}$	1648	
		5.8
← $\text{Ta}_3\text{Al}$	1352	
← $\text{Ta}$ Edge	229	
Center	192	4.9
TOTAL		16.9

Plate No. 23666

500X

Sample G14B19-1/2PC191-51

Fig. 9c. Oxidized 96 hours at 2500°F.



← $\text{Al}_2\text{O}_3$		0.46
← $\text{Ta}_2\text{Al}$	1258	
		3.5
← $\text{Ta}_3\text{Al}$	1250	
← $\text{Ta}$ Edge	196	
Center	202	5.6
TOTAL		13.8

Plate No. 23709

500X

Sample G14A19-1/2PC191-29

Fig. 9c. Oxidized 10 hours at 2700°F.

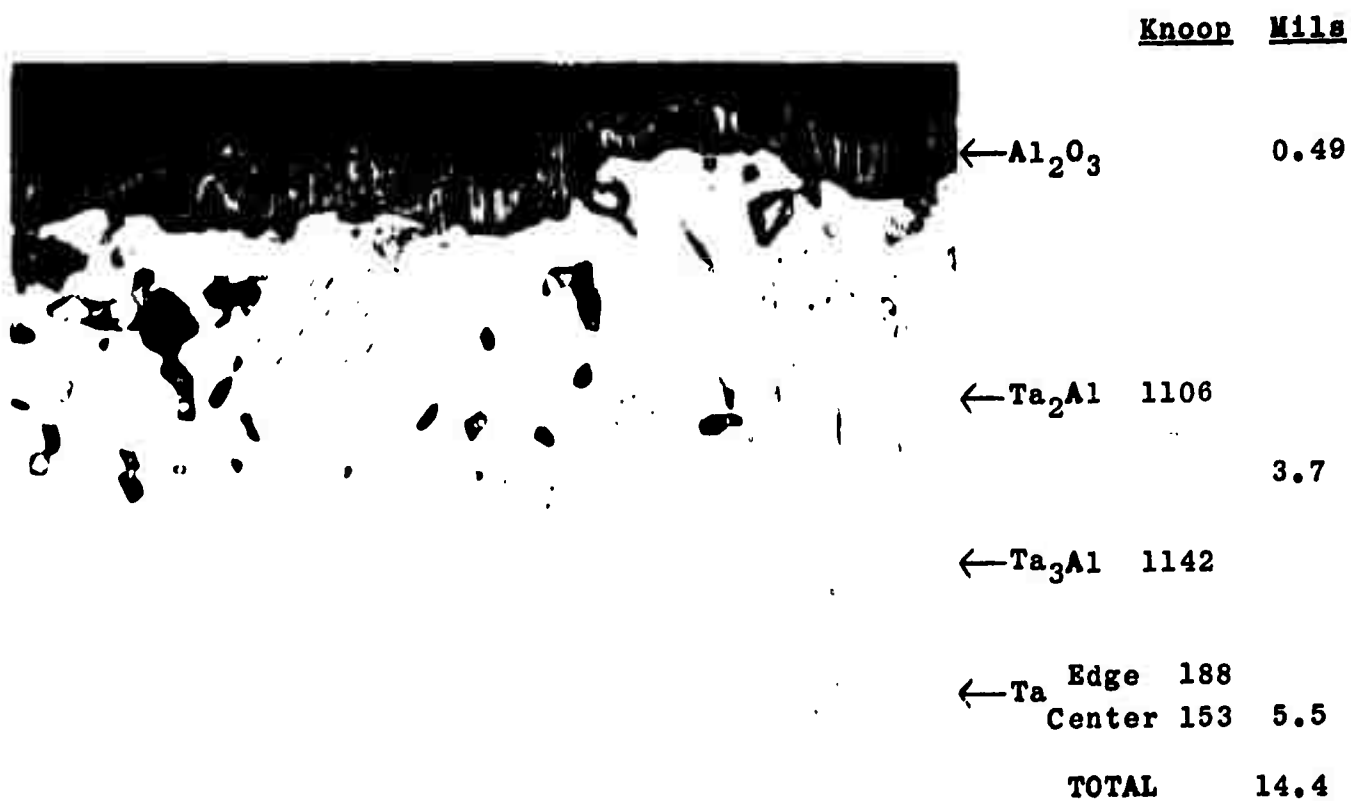


Plate No. 23707

500X

Sample G14191PC191-20

Fig. 9d. Oxidized 10 hours at 2800°F.

ten hours at 2800°F have only reacted with 2 to 2.5 mils per side. Microhardness tests indicate some internal hardening although it does not appear too serious. The maximum core hardness increases from 130 KHN for the as-received cold worked tantalum to 230 KHN after oxidation. The effect of oxidation time at 2500°F on reaction rates and hardness is plotted in Figure 10. Metallographic examination revealed no evidence of internal second phase precipitation.

### III. TIN-ALUMINUM COATINGS

#### A. Dip Coating

Coating thickness can usually be controlled by adjusting temperature and time but, with 1900°F or so, dipping temperature times are in seconds. To obtain better control of time, tin was added to the aluminum alloy compositions to act as a diluent and reduce the aluminum concentration. The first composition tried was 90 Sn-10 (Al-11 Si) (G9). Dipping time was 10 minutes at 1900°F following by a 1 hour pack calorizing heat treatment at 1900°F. After this processing schedule the coating was 2 mils (0.002") thick per side, 1 mil of  $TaAl_3$  and 1 mil of unreacted Sn-Al alloy, with only 0.5 mil core regression per side. Shorter dipping times gave a nonuniform coating thickness with variable oxidation results. Longer dipping times at lower temperatures did not give any better results, in fact poorer. Figure 11 shows the microstructure of an oxidized sample.

Silicon additions to aluminum compositions did not improve oxidation behavior. Therefore, the 90 Sn-10 Al (G21) composition was tried next. The effect of diffusion heat treatment is given in Figure 12. This coating on tantalum will withstand considerable abuse, in fact it shows some self-healing abilities. Tensile samples have been abraded into the tantalum core, Vickers hardness impressions put into the composites, twisted, bent 90° over a 0.020" die radius, in fact twisted into a knot and then electrically heated to 2600°F for 10 hours with 10 cycles without oxidation failure. A picture of the knot sample after test is given in Figure 13.

Using tensile type direct resistance heated samples for oxidation testing, the following statements can be made about the G9 and G21 compositions:

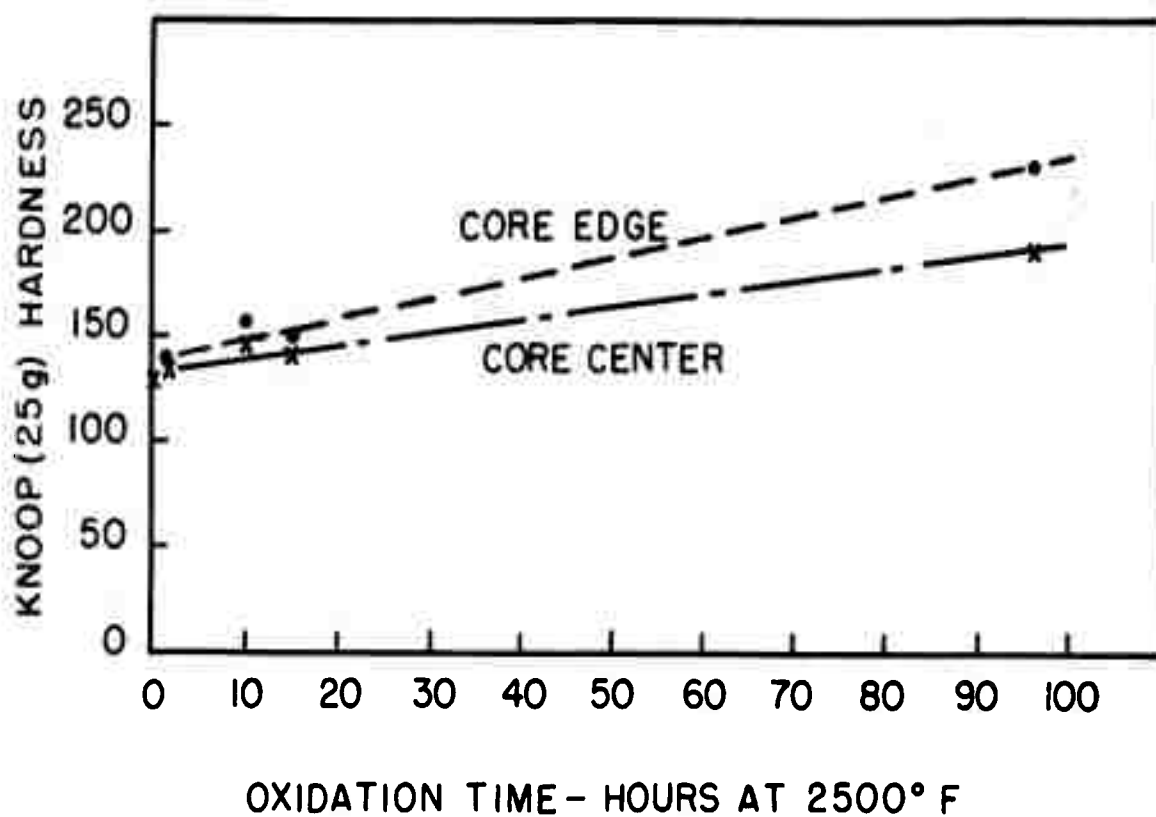
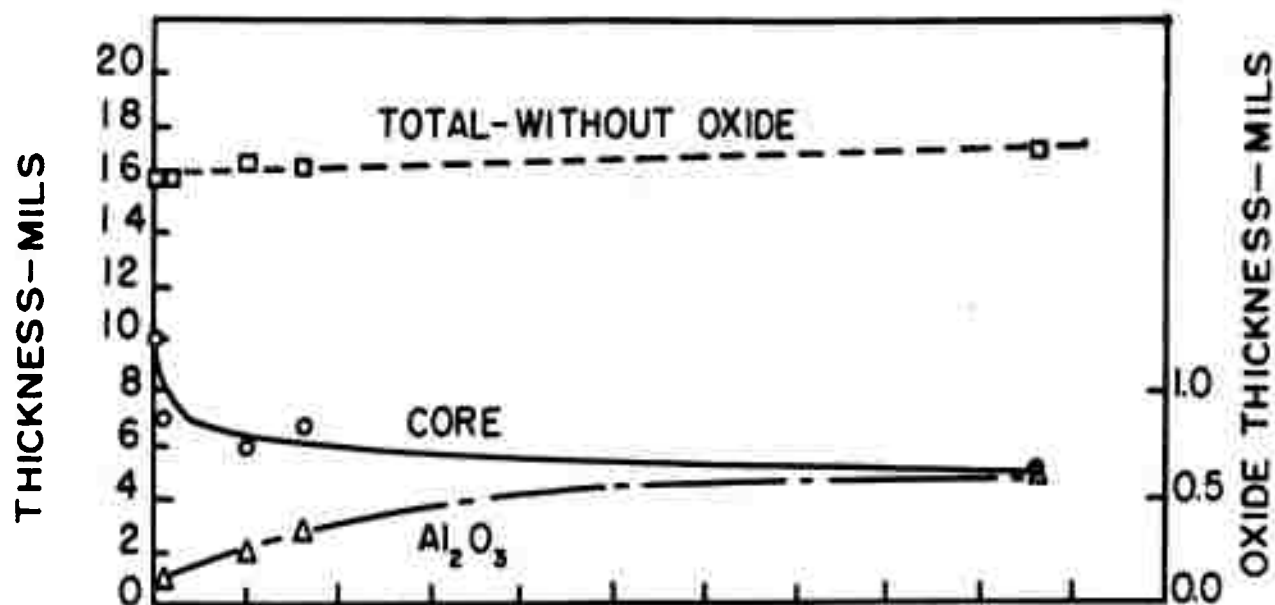


Fig. 10. Effect of time on core thickness and internal hardening of samples dip coated with Al-5 Cr-5 Ti.





	<u>Knoop</u>	<u>Mils</u>
← Coating	1101	1.2
← Ta		
Edge	126	
Center	123	7.9
TOTAL		11.8

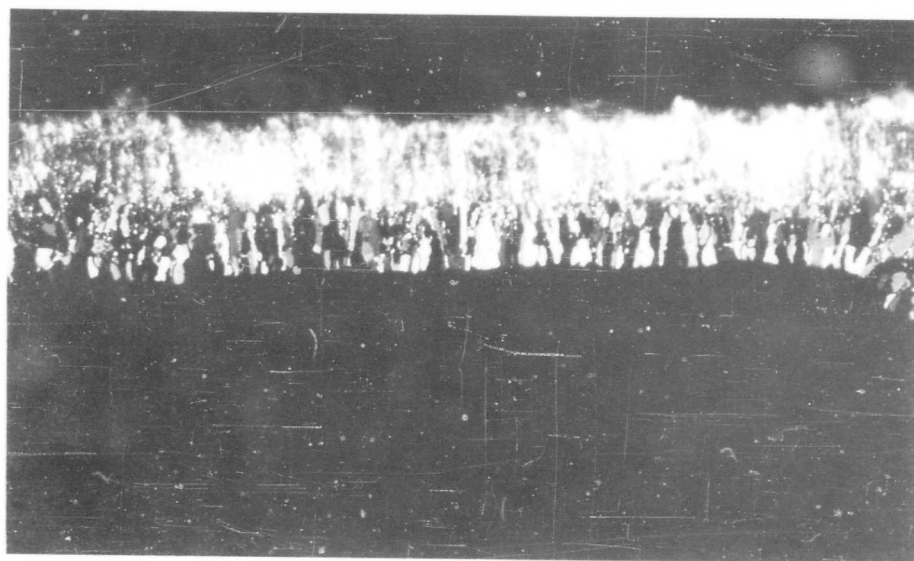
Plate No. 23678

500X

Sample G91910PC191-21

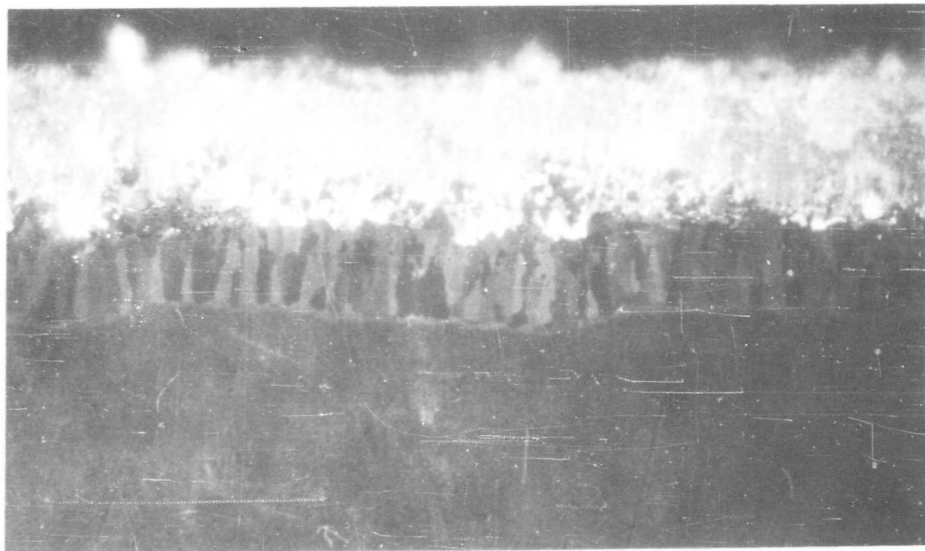
Bright field.

Fig. 11. 90% tin-10% (Al-10 Si) coated tantalum after oxidized 10 hours at 2600°F.



	<u>Knoop</u>	<u>Mils</u>
←Unreacted		1.2
←TaAl <sub>3</sub>	609	1.2
←Ta Edge	117	
Center	114	8.9
TOTAL		15.4

Plate No. 23670      Sample G211910PC191      500X  
1 hour 1900°F



←Unreacted		1.3
←TaAl <sub>3</sub>		
←TaAl <sub>2</sub>	1034	1.0
←Ta Edge	117	
Center	115	9.7
TOTAL		14.2

Plate No. 23671      Sample G211910PC221      500X  
1 hour 2200°F

Fig. 12. Effect of diffusion temperature on 90% tin-10% aluminum coated tantalum.

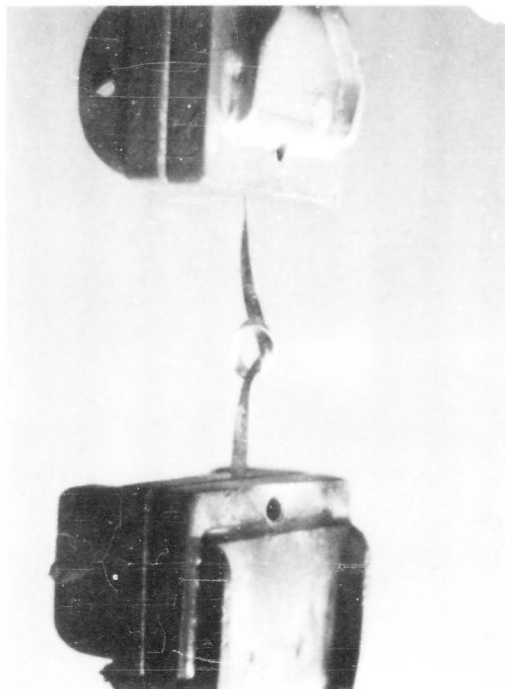


Fig. 13. 90% Sn-10% Al coated tantalum  
twisted into a knot.

1. Dipped and diffused samples will consistently withstand under stress ten hours and ten cycles (to sixty) to room temperature in an air blast at 2500° and 2600°F without any oxidation failure.
2. Samples were heated to 2700°F for ten hours without failure but this and higher temperatures seem to be beyond their capabilities since these compositions will not withstand thermal cycling as well as other compositions.
3. As-dipped samples of G21 will readily withstand ten hours and ten cycles at 2500° and 2600°F successfully. Diffusion treatment seems to be detrimental as compared to as-dipped samples.

Notwithstanding these results, it is important to note that samples using G9 or G21 will not take furnace oxidation at 2500°F for ten hours with cycling to room temperature. G21 coated Ta-10 W alloy samples would consistently withstand 16 hours isothermally at 2500°, 2600°, and 2700°F in a furnace. Most samples fail in 2 to 7 hours with hourly cycles to room temperature. Cyclic failure was probably caused by reaction of the coating with the ceramic boats. The effect of oxidation time at 2500°F on reaction rates and hardness is given in Figure 14. The Ta-10 W alloy recrystallized during oxidation causing a softening effect. The decrease in hardness of the Sn-Al phase was caused by the reaction of Al with Ta, reducing the Al concentration to a minimum and the hardness to that of almost pure tin. No hardness increase was found after oxidation with the pure tantalum core. Lower temperature oxidation behavior of these tin-aluminum coatings was better than the tin-free aluminum compositions. It was good enough for ten hour service but still not satisfactory by the arbitrary standards imposed here.

Different ratios of tin and aluminum were tried. A molten dip of 99 Sn-1 Al would not wet the tantalum surface to produce a  $TaAl_3$  coating. At least 3 Al was needed to wet the surface. The 95 Sn-5 Al bath (G50) would give the proper coating with no gain in oxidation resistance. Coating behavior was different when the Sn-Al powders were painted on, followed by a vacuum diffusion heat treatment for 1/2 to 1 hour. A 90 Sn-10 Al powder mix (G26 and G35) would not wet satisfactorily. The 50 Sn-50 Al alloy coatings (G34 and G36) did wet all surfaces of either pure Ta or the Ta-10 W alloy. Silver and copper powders have been substituted for tin to act as the diluent (G42 and G49) and the Cu-Al composition shows

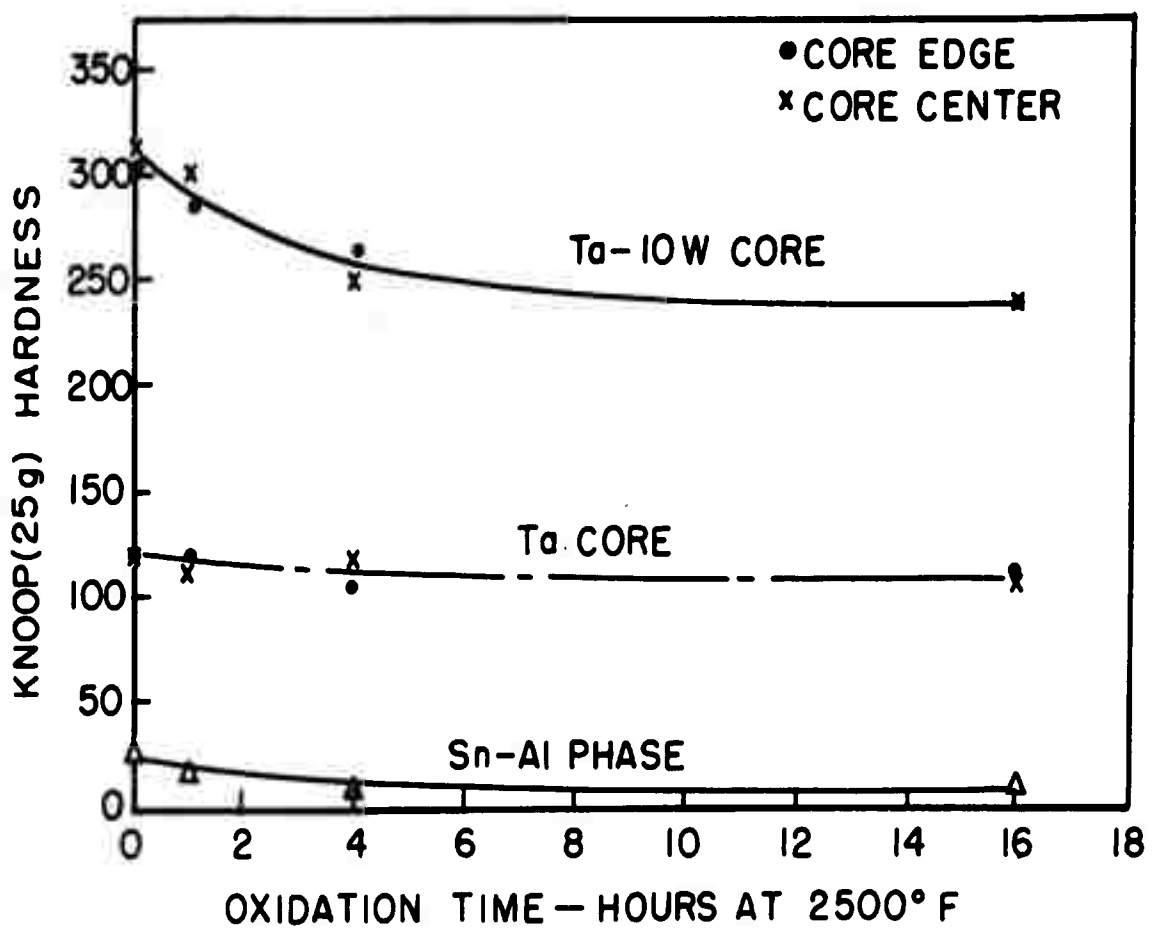
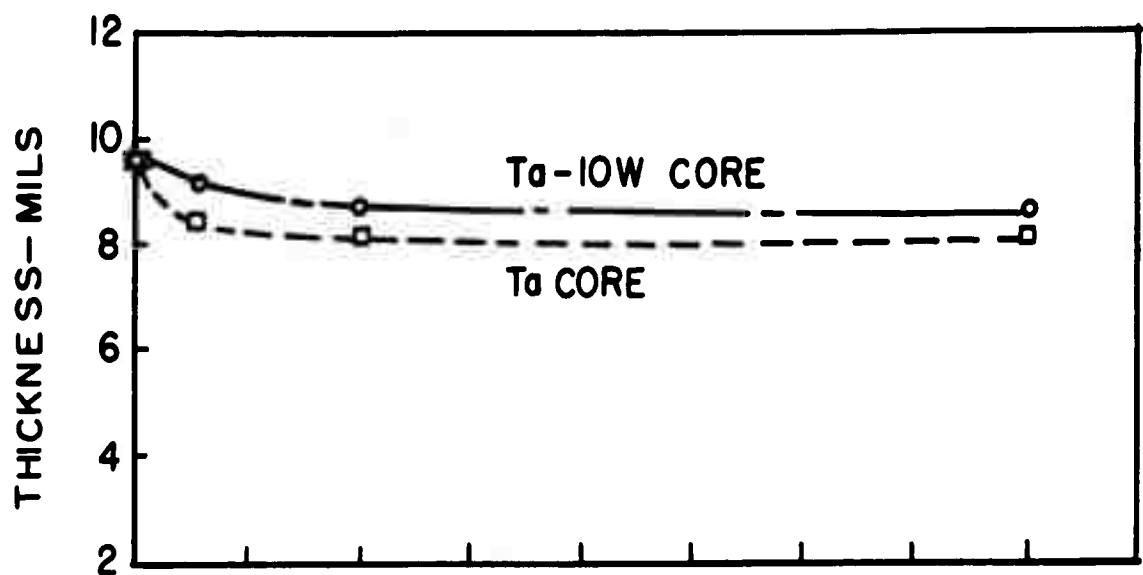


Fig. 14. Effect of time on core thickness and internal hardening of samples dip coated with Sn-10 Al.

enough merit to warrant more work. One advantage that copper has over tin is its solubility for other metals such as titanium and chromium.

## B. Spray Coating

Since dipping in a molten bath has many obvious practical limitations, other coating techniques were tried. Pack calorizing has already been discussed but no advantage could be seen by this process to produce tin-aluminum type coatings. Painting and spraying techniques look attractive. It is hard to visualize a simpler method. To concentrate effort on one composition the 50 tin-50 aluminum powder mixture using a nitrocellulose carrier was selected (G34). Preliminary oxidation results on painted and vacuum diffused samples, 1 hour at 1900°F, showed some promise but were not outstanding. During this time period initial work was started on spraying. The effect of a number of spray passes over the sheet sample on the coating thickness followed by a vacuum diffusion heat treatment after the final spray is given in Table 3. Furnace oxidation results at 2600°F on these samples are given in Table 4. Four passes give the best cyclic oxidation behavior. The microstructure of a sprayed sample is given in Figure 15. The optically active  $TaAl_3$  compound is visible on the Ta-10 W alloy substrate with a thin layer of Sn-Al on the outer surface. Extreme run-off of molten Sn-Al alloy occurs during vacuum diffusion heat treatment which agglomerates and remains on the sample whether applied by painting or spraying techniques. This either runs off and disappears when put into the oxidation furnace or flows back onto the coated surface. Evidence of both has been detected. The samples are suspended vertically during heat treatment. Apparently surface tension holds enough coating alloy on the tantalum surface to form the required two mil coating of  $TaAl_3$ .

The next logical step was to spray with one pass of the spray gun, react by vacuum diffusion for 1/2 hour at 1900°F, respray and rediffuse. For simplicity this coating procedure is identified as 348. Microstructures of an as-coated sample and one that had been oxidized for 10 hours with 9 cycles at 2700°F in a furnace without failure are shown in Figure 16. The edge of the as-coated sample shows how  $TaAl_3$  grows perpendicular to the alloy surface leaving a large void which the Sn-Al alloy fills in. All samples of either pure tantalum or Ta-10 W alloy oxidized for any time or temperature above 2000°F look exactly alike. The voids in the  $Ta_2Al$  are filled with essentially pure tin with a dense impervious outer  $Al_2O_3$  layer. Thermally cycled samples have the rough surface as shown by the

TABLE 3  
THICKNESS OF SPRAYED AND DIFFUSED Sn-Al  
COATING ON Ta-10 W ALLOY

<u>Passes</u>	<u>Thickness in mils (0.001")</u>			<u>Total</u>
	<u>Coating</u>	<u>Core</u>	<u>Coating</u>	
1	2.1	9.1	2.1	13.3
2	2.1	8.9	2.1	13.1
3	2.2	8.9	2.2	13.3
4	2.5	8.7	2.5	13.7

Samples were vacuum diffused for .1 hour at 1900°F after the number of passes.

TABLE 4

FURNACE OXIDATION RESULTS ON MULTI-SPRAYED  
Sn-Al COATED Ta-10 W ALLOY AT 2600°F

<u>Passes</u>	<u>Time, Hours</u>	<u>Cycles</u>	<u>Remarks</u>
1	16		Good
2	<16		Failed
3	16		Good
4	16		Good
1	2	1	Failed
2	2	1	Failed
3	3	3	Failed
4	8	7	Good





Plate No. 23941

Polarized light

500X

Sample 4TG34SVD191

Fig. 15. Spray coated four times and vacuum diffused sample of Ta-10 W alloy.

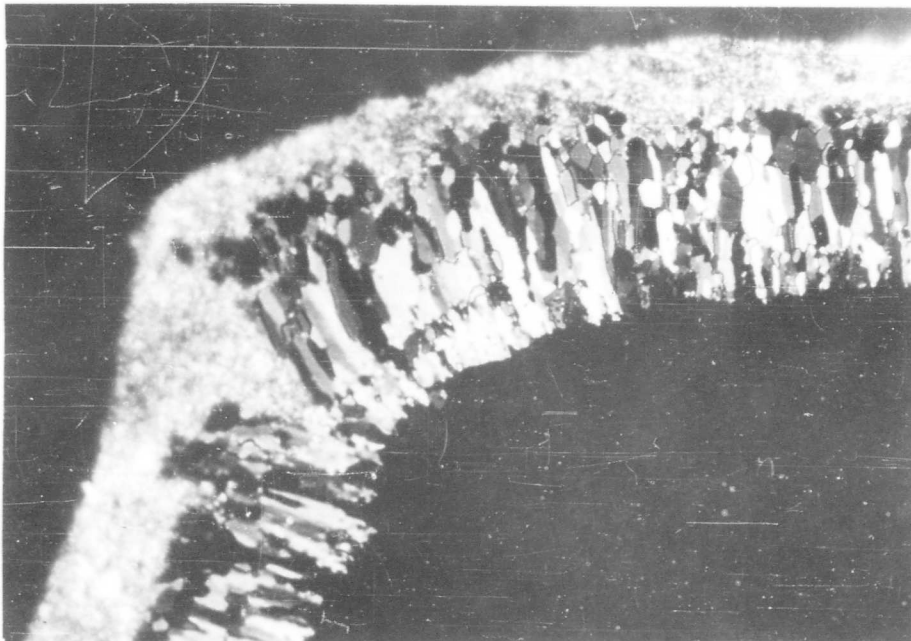


Plate No. 24025      Polarized light      500X  
Sample (TG34SVD19-1/2)<sup>2</sup>

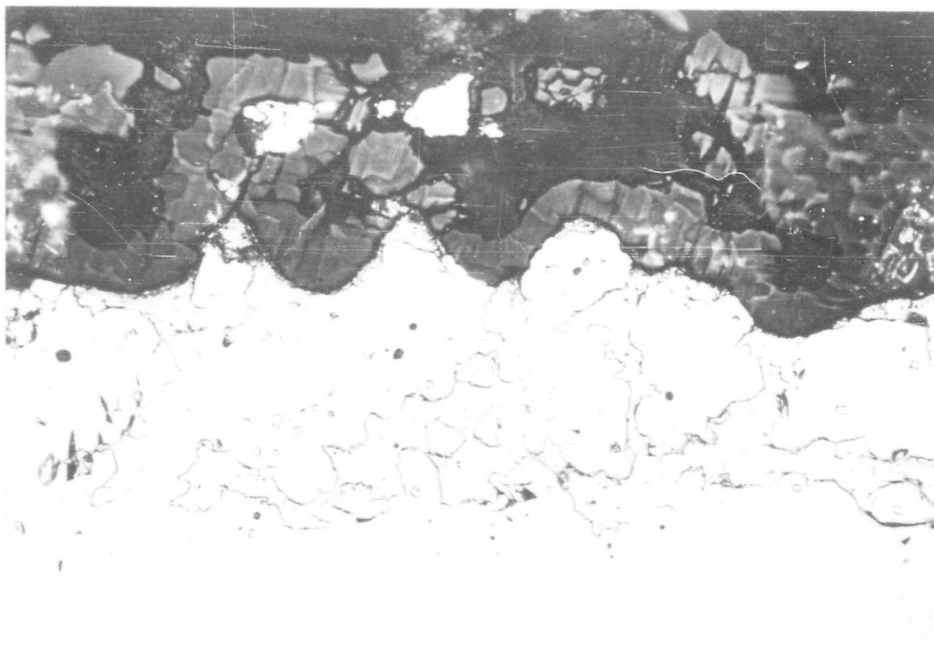


Plate No. 24026      500X  
Sample (TG34SVD19-1/2)<sup>2-23</sup>

Fig. 16. As coated and oxidized 10 hours at 2700°F.  
Sample of (G34S)<sup>2</sup> Ta-10 W alloy.

oxide contour. This is caused by the different thermal expansion coefficients of the various components and the fact that the molten tin can relieve strains until it freezes at about 450°F. Samples oxidized isothermally have a smooth oxide surface.

All of the furnace oxidation test results using the 34S composition are given in Table 5. This coating system meets all of the requirements of oxidation protection for 10 hours with a minimum of 10 thermal cycles in the temperature range of 2500°-2800°F quite easily. A more reasonable evaluation is 100 hour life at any temperature or any combination of temperatures between -320°F to 2800°F for the Ta-10 W alloy. Oxidation results up to 2950°F are also noteworthy. Coated samples were cooled to -320°F for 1 hour, oxidized for 4 hours at 2800°F, kept at -320°F for 16 hours and reoxidized for 4 hours at 2500°F without failure or any formation of "tin pest".

#### C. Battelle Alloys

Battelle Memorial Institute has developed two tantalum base alloys which have certain advantages. One has lower density, Ta-30 Cb-7.5 V, and the second, Ta-10 Hf-5 W, has good elevated temperature properties. The first alloy was produced in button form at Sylcor and cold rolled to 10 mil thick sheet. Coated samples gave poor oxidation results which are given in Table 6. This alloy reacts faster with the aluminum than other tantalum materials and behaves more like a columbium alloy.

The second alloy was received from Battelle and cold rolled to 10 mil thick sheet. The furnace oxidation results are also listed in Table 6. Initial work using the 34S coating revealed an unknown added phase next to the core material. Reaction rates are faster also. These coated alloys will be discussed further in the next section.

#### D. Bend Ductility and Diffusion Rates

To determine diffusion rates and effect of oxidizing conditions on bend ductility of coated tantalum materials, coupons of 10 and 25 mil thick pure tantalum sheet and 10 mil thick sheet of the Ta-10 W alloy and the two Battelle alloy compositions were coated with G21 and 34S. Samples were oxidized for 1, 5, and 16 hours at 2500° and 2600°F. It was found that the coated Ta-10 W alloy would not take a bend without cracking through. This is indicative of a highly notch sensitive alloy since no internal hardening and little

TABLE 5

## FURNACE OXIDATION RESULTS ON 348 COATED Ta-10 W ALLOY

<u>Sample</u>	<u>Time, Hours</u>	<u>Cycles</u>	<u>Remarks</u>
<u>2960°F Tests</u>			
34	3		Good
162	6	5	Good
163	6	5	Good
36	8	7	Good
Flame sample	10	9	Failed at Hole
35 and 35A	16		Good
130	64		Good
131	64		Failed
158	65		Good
159	65		Good
<u>2800°F Tests</u>			
26	6	5	Failure Due to Mishandling
25	8	7	Failure Due to Mishandling
70	10	9	Good
71	23	22	Good
38	>40 <44	3	Failed
54	52	6	Failed
55	56	7	Good
42	64		Good
67	65		Good
39	>80 <96	7	Failed
77	112		Good
<u>2700°F Tests</u>			
23	10	9	Good
24	10	9	Corner Failure
27	<16		Failed
28 and 29	16		Good
66	63	7	Failed
30 and 31	64		Good
52	77	9	Failed

TABLE 5 (Cont'd.)

## FURNACE OXIDATION RESULTS ON 348 COATED Ta-10 W ALLOY

<u>Sample</u>	<u>Time, Hours</u>	<u>Cycles</u>	<u>Remarks</u>
<u>2700°F Tests (Cont'd.)</u>			
53	80	10	Failed
64	95	6	Failed
65	110	7	Failed
<u>2600°F Tests</u>			
22	5	4	Good
21	16		Good
24	16		Good
62	100	10	Failed
63	100	10	Failed
68	120	12	Failed
69	120	12	Failed
<u>2500°F Tests</u>			
20	16		Good
49	80	2	Failed
50	92	3	Failed
32	112		Good
40	152	8	Failed
94	188	8	Good
56	208	20	Failed
41	216	13	Corner Failure
57	256	23	Failed
51	368	30	Failed
<u>2200°F Tests</u>			
47	136	9	Failed
48	240	14	Edge Failure
58	288	23	Good
59	288	23	Failed

TABLE 5 (Cont'd.)

## FURNACE OXIDATION RESULTS ON 348 COATED Ta-10 W ALLOY

<u>Sample</u>	<u>Time, Hours</u>	<u>Cycles</u>	<u>Remarks</u>
<u>1800°F Tests</u>			
45	528	21	Good
46	528	21	Good
<u>1400°F Tests</u>			
43	528	21	Good
44	528	21	Good

TABLE 6

## FURNACE OXIDATION RESULTS ON COATED BATTELLE ALLOY SHEET

<u>Coating</u>	<u>Time, Hours</u>	<u>Cycles</u>	<u>Remarks</u>
<b>Ta-30 Cb-7.5 V Alloy</b>			
<u>2500°F Tests</u>			
G21	8	1	Good
G21	<16		Failed
G48 with G34 overlay	10	9	Good
<u>2600°F Tests</u>			
G21	8	1	Good
G21	16		Good
G21	5	4	Failed
G21	<16		Failed
G48 with G34 overlay	<16		Failed
<u>2700°F Tests</u>			
G21	3	2	Failed
G48 with G34 overlay	<7		Failed
<b>Ta-10 Hf-5 W</b>			
<u>2500°F Tests</u>			
36S	38	12	Good
36S	64	6	Through Core
			Coating Diffused
34S	10	10	Good
34S	38	12	Good
34S	88	10	Failed
<u>2700°F Tests</u>			
36S	<16		Failed
34S	16	2	Failed

diffusion takes place during coating. A sample of the alloy with the same heat treatment was found to bend without any change in hardness, 285 VPN. Bend ductility tests indicated little harmful effect of internal contamination with the G21 coating on 10 mil tantalum. They were ductile in a room temperature bend of 90° over a 4T radius after up to 16 hours exposure at 2500°F and 2600°F. The coating itself cracked but the crack did not propagate appreciably into the substrate. This is shown in Figure 17.

Reaction rates are given in Table 7 for most of the samples with the G21 coating. Thickness data with the G21 coating show little significant difference between Ta or Ta-10 W alloy. The metal reacts with all of the available aluminum with no additional decrease in core thickness. The effect of excess aluminum in the 34S composition with the three tantalum materials is given in Table 8. With most oxidization conditions the Ta-10 Hf-5 W alloy had no core left. The harmful effect of the run-off remaining on the samples is evident also. The area of the excess Sn-Al has reacted with all of the Ta-10 W alloy core in certain cases. Measurements were made at the top of the sample, middle, and the bottom area with the run-off. Obviously the excess must be removed before test. This can be done quite easily depending on the geometry of the sample. One way is to heat the sample to a black heat, enough to melt tin (450°F), and shake or wipe it off.

#### E. Oxidation Rates

Prior work has shown that the 34S coating would meet the proposed oxidation requirements and it was felt desirable to obtain weight gain data as a function of time and temperature. Tests were conducted on coated Ta-10 W alloy at 1800°, 2200°, 2500°, and 2800°F. The results are listed in Table 9 and plotted in Figure 18. Thermal cycling accelerates oxidation by a factor of two or more. At 1800° and 2200°F the samples lose weight because of the run-off of excess Sn-Al. But at 2500° and 2800°F no loss in weight occurs, indicating that the excess coating runs back up onto the surfaces. With this coating, weight gain data gives no indication of protectiveness since the Ta-10 Hf-5 W alloy oxidizes at a lesser rate than the Ta-10 W alloy but has markedly poorer oxidation resistance; it fails sooner, presumably because of rapid diffusion.



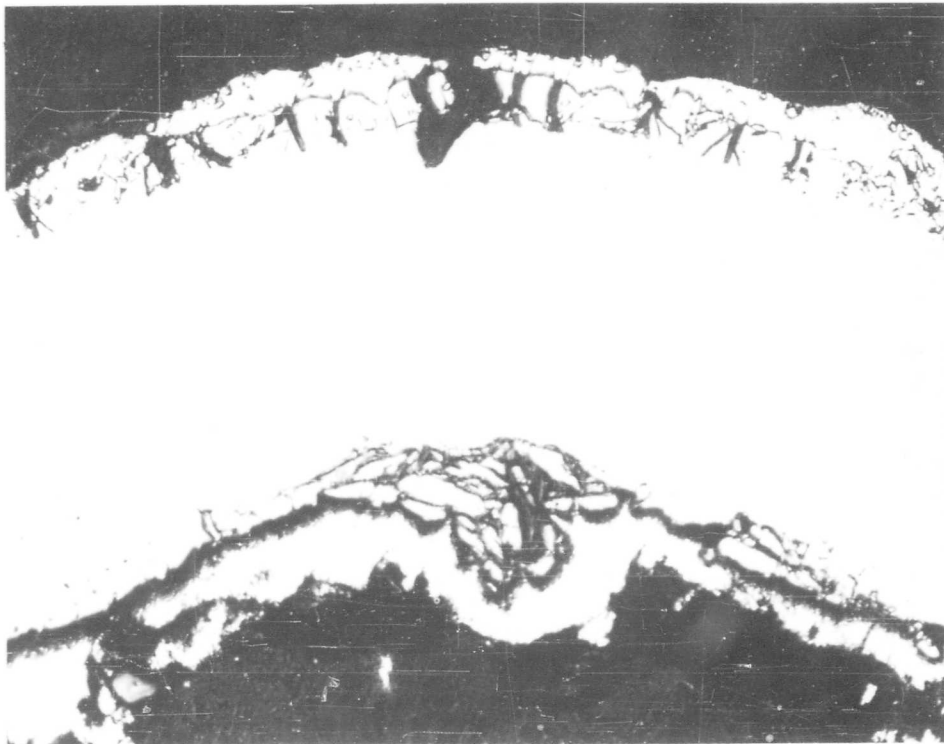


Plate No. 23985

4T bend in Ta

200X

Fig. 17. Overall view of bend area, oxidized sixteen hours at 2600°F - Coating Sn-10 Al dip.

TABLE 7

## DIFFUSION EFFECTS ON G21 COATED TANTALUM MATERIALS

<u>Thickness in mils (0.001")</u>			
<u>Tantalum</u>	<u>Ta-10 W</u>	<u>Ta-10 Hf-5 W</u>	<u>Ta-30 Cb-7.5 V</u>
<u>As-Dipped</u>			
Coating	1.6	1.8	1.3
Core	9.7	9.7	9.6
Coating	<u>1.6</u>	<u>2.0</u>	<u>1.2</u>
Total	12.8	13.5	12.1
<u>1 Hour 2500°F</u>			
Coating	2.1	2.9	2.8
Core	9.2	8.4	7.9
Coating	<u>2.3</u>	<u>3.6</u>	<u>2.7</u>
Total	13.6	14.9	13.4
<u>5 Hours 2500°F</u>			
Coating	2.7	2.8	2.6
Core	8.7	8.1	7.4
Coating	<u>2.5</u>	<u>2.6</u>	<u>2.7</u>
Total	13.9	13.5	12.7
<u>16 Hours 2500°F</u>			
Coating	2.6	3.3	
Core	8.7	8.1	Failed
Coating	<u>2.5</u>	<u>3.5</u>	
Total	13.8	14.9	
<u>1 Hour 2600°F</u>			
Coating	2.5	2.8	
Core	9.3	8.4	Failed
Coating	<u>2.9</u>	<u>2.9</u>	
Total	14.7	14.1	

TABLE 7 (Cont'd.)

## DIFFUSION EFFECTS ON G21 COATED TANTALUM MATERIALS

<u>Thickness in mils (0.001")</u>			
<u>Tantalum</u>	<u>Ta-10 W</u>	<u>Ta-10 Hf-5 W</u>	<u>Ta-30 Cb-7.5 V</u>
<u>5 Hours 2600°F</u>			
Coating	2.5	3.6	
Core	8.7	8.4	
Coating	<u>2.8</u>	<u>3.4</u>	
Total	14.0	15.4	
<u>16 Hours 2600°F</u>			
Coating	3.3	2.6	2.5
Core	8.4	8.2	6.7
Coating	<u>3.8</u>	<u>2.8</u>	<u>2.2</u>
Total	15.5	13.6	11.4

# DIFFUSION EFFECTS ON 348 COATED TANTALUM MATERIALS

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TABLE 8 (Cont'd.)

## DIFFUSION EFFECTS ON 348 COATED TANTALUM MATERIALS

		Thickness in mils (0.001")					
		Tantalum			Ta-10 W		
		Top	Center	Bottom	Top	Center	Bottom
		1 Hour 2600°F					
Coating	7.4	9.2	9.9	10.8	12.4		
Core	20.4	19.8	18.6	3.0	2.0		
Coating	8.7	9.9	11.0	11.6	12.4		
Total	36.5	38.9	39.5	25.4	26.8		37.7
		5 Hours 2600°F					
Coating	11.6	12.4	12.6	7.4	8.9		11.2
Core	19.8	18.6	16.0	6.0	4.9		1.2
Coating	9.9	11.9	12.6	5.7	8.8		12.4
Total	41.3	42.9	41.2	19.1	22.6		24.8
		16 Hours 2600°F					
Coating	7.4	12.4	12.4	16.8	11.9		
Core	19.8	15.8	14.8	2.5	1.1		
Coating	7.2	12.4	13.4	11.6	11.9		
Total	34.4	40.6	40.6	30.9	24.9		24.8

TABLE 9

## OXIDATION WEIGHT CHANGE OF 348 COATED Ta-10 W ALLOY

Time, Hours	Temperature, °F					
	Cycled				Isothermal	
	1800	2200	2500	2800	2500	2800
<u>Weight Change in Milligrams</u>						
1	-5.1	-0.5	3.2	6.6		
2	-2.9	-0.8	5.8	9.8		
4	-18.2	-0.3	8.5	16.1		
8	Failed	0.2	16.2	27.3		
24		0.8	34.1	42.4		
46			43.0	62.5		
70			47.6	Failed		
100					18.1	37.9

Ta-10 Hf-5 W Alloy

1	2.3
2	3.9
4	7.6
8	10.3
24	20.3

Sample Size - 0.5" x 0.75" x 0.10" uncoated.  
 Coated Weights - 1.2 to 1.5 grams.

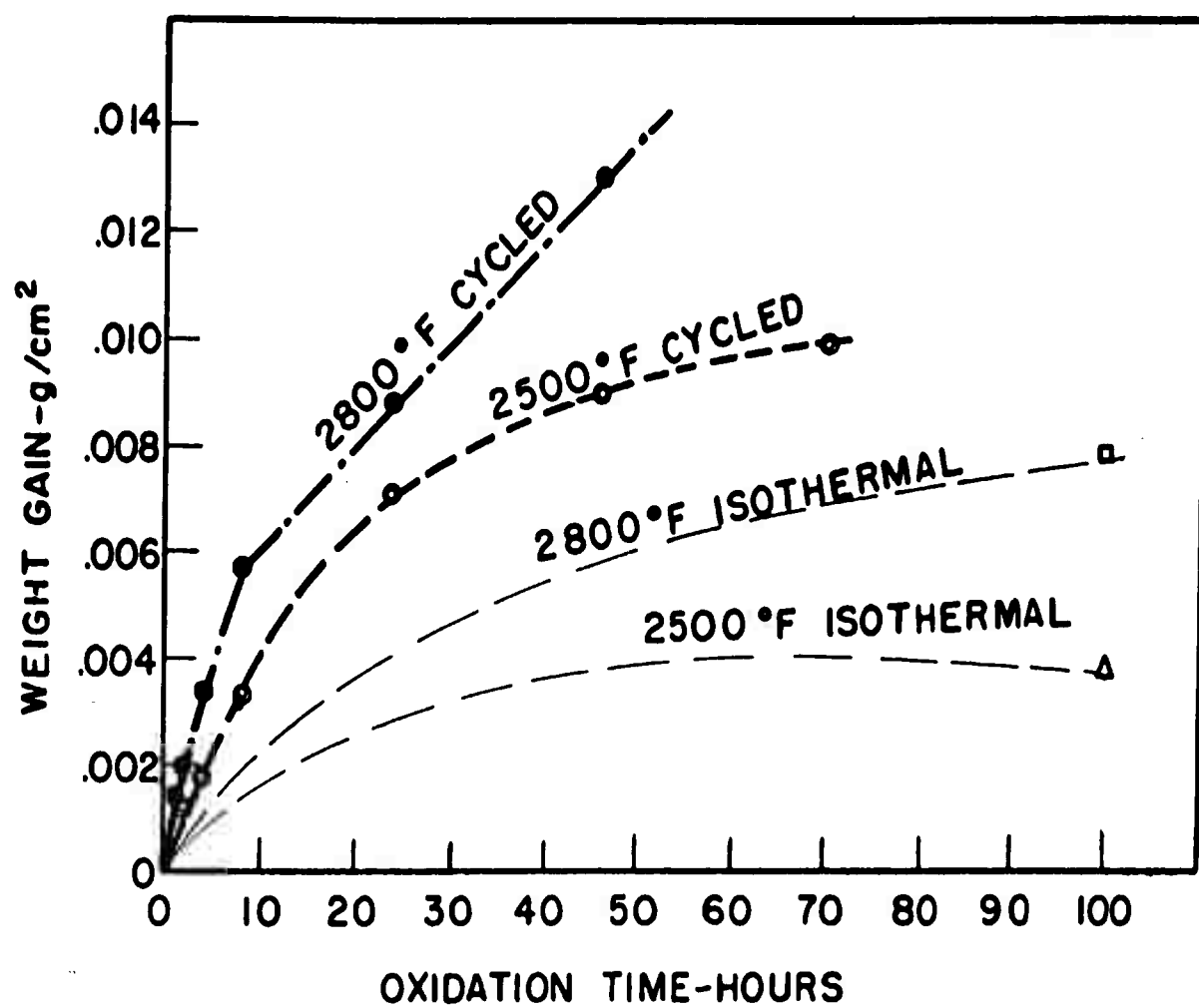


Fig. 13. Weight gains of 34S coated Ta-10 W alloy samples oxidized at 2500° and 2800°F.

#### F. Electrically Heated Oxidation Data

Direct resistance heating was used to evaluate many coating compositions. Two machines were powered by DC current and two by AC current. At first no difference in oxidation results was attributed to AC or DC current. Many samples oxidized and failed in the cooler regions and were classified as cold zone failures, as opposed to those that failed in the hot zone. This could be justified by the poor low temperature oxidation properties of aluminum compositions. But when testing Sn-Al compositions too many duplicate tests were failing at short times in the cold zone. Samples also appeared to be embrittled. The effect of DC and AC power on 348 coated Ta-10 W samples is evident in Table 10. A "knuckle" of material is formed at the negative electrode in DC tests no matter if it is at the top or bottom of the fixture and premature failure occurs at this area. Mass transfers occur by electrolytic action. All of the Ta alloy reacts with Al to form  $Ta_2Al$  across the entire cross-section forming the "knuckle". Therefore only AC heated results are valid. These tests do show that the 348 coating can withstand up to 100 cycles in the temperature range of 2500°-2800°F.

#### G. Creep Oxidation

To get a better engineering viewpoint with respect to size effects and ability to withstand mechanical stress and resultant strain on coated samples, larger creep samples were fabricated, coated, and oxidation tested. The specimens were welded together using 6" long strip of the Ta-10 W alloy with a final size of 0.010" thick by 1" wide x 18" long. Tabs were spot welded along the side for creep measurement purposes. Holes were punched in the cold ends for gripping. The creep results obtained are listed in Table 11. The coating on the first eight samples was painted on, the balance were sprayed. More extensive work needs to be done in this area. Although a few samples would last 10 hours at temperature with more than 2% strain, different results were obtained in different furnaces. A better explanation for the random results is that a larger vacuum furnace had to be used for diffusion heat treatments. The heating rate is slower, which could have a harmful effect. Actually no explanation can be given at the present time.

#### H. Flame Oxidation Tests

Another screening test used to evaluate coating, especially at temperatures difficult to obtain by more conventional means, was



TABLE 10

DIRECT RESISTANCE HEATED OXIDATION RESULTS  
ON 348 COATED Ta-10 W ALLOY

<u>Sample No.</u>	<u>Temperature, °F</u>	<u>Time, Hours</u>	<u>Cycles</u>	<u>Power</u>	<u>Remarks</u>
76A	2500	10	80	DC	BOR
75	2500	4.8	38	DC	CZF
92	2700	1	1	DC	CZF
83	2700	1.2	12	DC	CZF
84	2700	1.2	12	DC	CZF
87	2700	1.4	2	DC	CZF
90	2700	1.6	3	DC	CZF
89	2800	0.8	1	DC	CZF
74	2500	10	100	AC	G
82	2600	10	104	AC	G
88	2700	10	20	AC	G
91	2700	10	54	AC	G
33	2800	10	13	AC	BOR
95	2800	10	60	AC	G
37	3000	2	2	AC	G

G - Sample good.  
BOR - Broke on removal.  
CZF - Cold zone failure.

TABLE 11

## CREEP RESULTS ON COATED Ta-10 W ALLOY

<u>Sample</u>	<u>Time, Hours</u>	<u>Cycles</u>	<u>Stress, psi</u>	<u>Elongation, %</u>	<u>Remarks</u>
<u>2500°F Tests</u>					
<u>G 34 Coating</u>					
SR 1	9		1300		Grip Broke
SR 2	6	1	5700	5	Failed
SR 3	9.5		4300	6	Good
SR 4	10		4300	2	Good
SR 5	6.5	3	4300	9	Failed
SR 6	7	3	2500	0.3	Failed
SR 9	6.7	3	2800		Failed
SR 10	0.5		3200		Failed
SR 11	5	3	2500		Failed
SR 12	6	3	2700		Failed
<u>G 22 Coating</u>					
SR 7	8.5	4	2000	2	Failed
SR 8	1.5		2100	0.4	Failed
<u>34S Coating</u>					
SR 13	10	5	2700		Good
SR 14	7	3	2400		Failed
SR 15	10	10	2600		Good
SR 16	8	7	3100		Grip Broke
<u>2600°F Test</u>					
<u>34S Coating</u>					
SR 17	8.7	4	1800		Failed

the flame test. At first a curved sample was used with the flame from a small oxidizing oxy-acetylene torch impinging against the convex surface. With our coatings it was felt that using a flat sample, 1" x 2" x 0.010", and a multi-flame torch that a larger area would be heated thereby giving a more representative result. The flame test can only be used to compare coating compositions relative to each other. The results obtained are given in Table 12. Once again thermal cycling causes more rapid failure as compared to isothermal conditions. Nevertheless certain coatings are capable of meeting the requirements of the program. Of more importance is the ability of Sn-Al compositions to withstand temperatures about 3000°F for practical time periods.

#### I. Arc-Plasma Tests

The 348 coating surpassed original requirements. The primary question was what would happen to this coating with its liquid second phase in potential service conditions? Of course the answer would eventually be found by testing it in hypersonic and/or space flight. A popular first approach to duplicating re-entry conditions is the arc-plasma jet. Therefore, samples of 348 coated Ta-10 W alloy were prepared and so tested. The test schedule and results are shown in Table 13. These serve to add further proof that the coating is protective in a relatively fast moving hot air stream to temperatures above 2800°F and is also resistant to thermal cycling. The cyclic results also indicate good reproducibility. The tests were run at the University of Dayton Research Institute and the test conditions were arbitrarily selected by them.

Several optical devices were used for temperature measurements and preliminary indications were that the coatings had a relatively high emissivity. However, no numerical values for this were given.

Comparison tests at Sylcor were made using an optical pyrometer and a Shawmeter, an instrument claimed to give true temperature regardless of emissivity, atmospheric conditions, etc. These tests also indicated an emissivity value close to unity for the oxidized 348 coating. This is another area that requires more study.

#### IV. COLUMBIUM OXIDATION RESULTS

A secondary part of this investigation was to determine how the better tantalum coatings performed when applied to columbium

TABLE 12

## FLAME OXIDATION TEST RESULTS ON COATED Ta-10 W ALLOY

<u>Sample</u>	<u>Time, Hours</u>	<u>Cycles</u>	<u>Temperature, °F</u>	<u>Remarks</u>
<u>Curved Samples</u>				
G21-2	8	8	2700	Good
G21-3	8	15	2700	Good
G21-4	4.7	5	2800	Failed
G21-5	2.7	3	3000	Failed
<u>Flat Samples</u>				
G21-10	8	8	2500	Good
G21-9	3.8	4	2700	Failed
G21-8	2.3	2	2800	Failed
G21-11	0.3	3	3000	Failed
G21-7	1	1	3000	Failed
G21-6	1.2	1	3000	Failed
G21-1	7.5		3000	Good
G21-5	7.7		3000	Good
G21-3	4.5	1	3200	Failed
G21-2	0.01		3400	Failed
G21-4	0.3		3400	Failed
G14-3	7.5	8	2600	Good
G14-1	10	10	2600	Good
G14-2	0.1		2800	Failed
G41-4	8	8	2500	Good
G41-3	1.7	1	2800	Failed
G41-2	0.3		3000	Failed
G41-1	1.7	1	3000	Failed
G47-4	6.5	7	2500	Failed
G47-3	1.5	1	2800	Failed
G47-1	0.5		3000	Failed
G47-2	1.5	1	3000	Failed

TABLE 12 (Cont'd.)

FLAME OXIDATION TEST RESULTS ON COATED Ta-10 W ALLOY

<u>Sample</u>	<u>Time, Hours</u>	<u>Cycles</u>	<u>Temperature, °F</u>	<u>Remarks</u>
<u>Flat Samples (Cont'd.)</u>				
G40 Paint-1	5.5	5	2700	Good
G40 Paint-2	4	4	3000	Good
G40 Paint-3	3	3	3000	Failed
G40 Paint-4	7.5	8	3000	Failed
G40 Spray-4	4.8	5	2500	Failed
G40 Spray-3	2.7	2	2800	Failed
G40 Spray-2	1	1	3000	Failed
G40 Spray-1	1	1	3000	Failed
40S-16	2	8	3000	Good
40S-18	0.15		3180	Failed
40S-17	0.5	2	3180	Failed
40S-11	1.7	3	3180	Failed
40S-12	2	2	3180	Failed
40S-13	1.3		3270	Failed
34S-1	8	8	2500	Good
34S-2	8	8	2600	Good
34S-3	8	8	2700	Good
34S-4	3.5	3	2800	Failed
34S-5	6.3	6	2800	Failed
34S-6	3.8	4	2900	Failed
34S	1	1	3000	Failed
34S	1.3	1	3000	Failed
34S-169	0.9	3	3000	Failed
34S-170	2	8	3000	Good
34S-168	0.1		3180	Failed

TABLE 13

## ARC-PLASMA TEST SCHEDULE ON 348 COATED Ta-10 W ALLOY

<u>Sample</u>	<u>Temperature, °F*</u>	<u>Time</u>	<u>Remarks</u>
1	1830	5 minutes	Temperature increased
	2540	5 minutes	Temperature increased
	2950	5 minutes	Temperature increased
	3230	5 minutes	No failure, test stopped
2	2550	5 minutes	Temperature increased
	2910	5 minutes	Temperature increased
	3260	5 minutes	Temperature increased
	3360	5 minutes	Full power, no failure
3	2730	Cycled**	15 cycles, temperature increased
	2910	Cycled**	15 cycles
	3150	Cycled**	4 cycles, failed
4	2730	Cycled**	15 cycles
	2910	Cycled**	5 cycles, failed
5	2730	Cycled**	15 cycles
	2910	Cycled**	5 cycles failed
6	2730	Cycled**	15 cycles
	2910	Cycled**	8 cycles failed
7	2730	Cycled**	15 cycles
	2910	Cycled**	8 cycles failed
8	2730	Cycled**	15 cycles
	2910	Cycled**	11 cycles failed

\* Uncorrected optical pyrometer readings.

\*\* A cycle is 30 seconds hot, 10 seconds cooled.  
Test samples 2" x 2" x 0.010" uncoated.  
Samples supported at 45° angle to hot air stream.

alloys. Oxidation results are summarized in Table 14. Once again the 348 system seems to be the best up to and including 2500°F. Around 2600°F and higher rapid reaction takes place between the Sn-Al coating and the substrate. 2500°F appears to be the maximum use temperature for Sn-Al coated columbium alloys.

## V. OTHER REFRACTORY METALS

The 348 coating was tried on molybdenum and tungsten. Arc-plasma, furnace, and flame test results are given in Table 15. Such promising indications were not entirely expected. Metallographic examination of the tungsten does reveal grain boundary attack. But this coating system requires and deserves much more development work since only a few samples were prepared merely for feasibility indications.

## VI. BERYLLIUM COATINGS

It is known that the tantalum beryllides are oxidation resistant at and above 2500°F<sup>(3)</sup>. The beryllides are: TaBe<sub>12</sub>, Ta<sub>2</sub>Be<sub>17</sub>, TaBe<sub>3</sub>, and TaBe<sub>2</sub>. Therefore, coatings were applied to the tantalum materials by vapor deposition techniques. The samples (0.5" x 0.75" x 0.010") were placed between powder rolled sheets of beryllium and vacuum heat treated. The beryllium sheet would sinter and could be used for many coating runs. The effect of coating conditions on thickness is given in Table 16. All four phases can be seen in the microstructure.

Early oxidation results on pure tantalum indicated that the coating was sensitive to thermal cycling since gross surface checking was noted on oxidized samples. It was apparent that beryllium coatings would confer the required oxidation resistance at 2500°F but that the difference in thermal expansion between the coating and the core should be matched more closely. Therefore, attempts were made to produce a lower beryllium content beryllide at the surface before oxidation testing. Lower beryllides should have lower coefficients of expansion more compatible with that of tantalum. Tantalum samples, 0.010" thick, were vapor coated by two different heat treatments, 25 hours at 1800°F and 4 hours at 2000°F. Half of each sample was then vacuum diffused for 25 hours at 2200°F. The change in the zone thickness is reported in Table 17. Unfortunately, the same furnace was used for diffusion as for coating and

TABLE 14

## OXIDATION RESULTS ON COATED COLUMBIUM ALLOYS

<u>Coating Treatment*</u>	<u>Temperature, °F</u>	<u>Time, Hours</u>	<u>Cycles</u>	<u>Remarks</u>
<u>Pure Columbium</u>				
8	1800	1000	30	Excellent
8	2300	148	5	Failed
8	2300	240	10	Failed
1 + 2	2300	31	15	Corner Failure
1 + 2	2500	13	13	Corner Failure
1 + 3	2300	48	17	Good
1 + 3	2500	24	24	Good
<u>B-22</u>				
8	1800	1000	30	Excellent
8	2300	112	4	Failed
8	2300	240	10	Failed
8	2500	100	12	Good
7	2500	10	9	Good
7	2500	24	1	Edge Failure
7	2500	28	27	Good
5	2300	96	4	Excellent
5	2500	38	38	Excellent
5	2500	64	4	Excellent
6	2300	96	4	Excellent
6	2500	38	38	Good
6	2500	64	4	Good
7	2500	10	9	Good
7	2500	23	22	Fair
7	2500	32	1	Fair
<u>F-48</u>				
1 + 2	2300	48	17	Fair
1 + 2	2500	13	13	Very Poor
1 + 4	2300	72	24	Good
1 + 4	2500	16	16	Poor
1 + 3	2300	72	24	Good
1 + 3	2500	16	16	Good



TABLE 14 (Cont'd.)

## OXIDATION RESULTS ON COATED COLUMBIUM ALLOYS

<u>Coating Treatment*</u>	<u>Temperature, °F</u>	<u>Time, Hours</u>	<u>Cycles</u>	<u>Remarks</u>
<u>F-48 (Cont'd.)</u>				
5	2300	120	4	Poor
5	2300	24		Poor
5	2500	7	7	Very Poor
6	2300	48	2	Very Poor
6	2500	32	2	Very Poor
6	2500	7	7	Very Poor

\* Coating Techniques

1. Titanized 16 hours at 2280°F in vacuum.
2. Pack calorized for 4 hours in argon at 1900°F in a powder mix of 40% (Al-11 Si-10 Cr)-2%  $\text{NH}_2\text{Cl}$ -58%  $\text{Al}_2\text{O}_3$ .
3. Dipped for 30 seconds at 1900°F in Al-10 Si-10 Cr and pack calorized for 1 hour at 1900°F in argon in 90%  $\text{Al}_2\text{O}_3$ -10% Al powders.
4. Dipped for 2 minutes at 1700°F in Al-11 Si and pack calorized.
5. Dipped for 15 seconds at 1900°F in Al-5 Cr-5 Ti and pack calorized.
6. Dipped for 15 seconds at 1900°F in Al-15 Cr-9 Si-5 Sn and pack calorized.
7. 36S.
8. 34S.

B-22 alloy is Cb-5 Hf-1 Ti-1 Zr, developed by Westinghouse under Air Force sponsorship.

TABLE 15

## OXIDATION RESULTS ON COATED MOLYBDENUM AND TUNGSTEN

<u>Coating</u>	<u>Type of Oxidation Test</u>	<u>Temp., of</u>	<u>Time</u>	<u>Cycles</u>	<u>Remarks</u>
<u>Molybdenum</u>					
34S	Arc-Plasma	2550	5 min.		Temperature increased
34S	Arc-Plasma	2910	5 min.		Temperature increased
34S	Arc-Plasma	3270	5 min.		Temperature increased
34S	Arc-Plasma	3450			Failed
34S	Arc-Plasma	2550	5 min.		Temperature increased
34S	Arc-Plasma	2910	5 min.		Temperature increased
34S	Arc-Plasma	3200	5 min.		No fail., test stopped
34S	Furnace	2500	46 hrs.	5	Good
34S	Furnace	2500	240 hrs.	11	Edge failure
34S	Flame	2730	20 min.		Failed
34S	Flame	2820	120 min.	4	Good
34S	Flame	2910	5 min.		Failed
34S	Flame	3000	5 min.		Failed
34S	Flame	3090	5 min.		Failed
34S	Flame	3180	5 min.		Failed
34S	Flame	3360	5 min.		Failed
34S	Flame	3270	33 min.	3	Failed
34S	Flame	3180	26 min.	5	Failed
34S	Flame	3000	10 hrs.	18	Good
40S	Flame	3300	68 min.	5	Good
40S	Flame	3400	51 min.	4	Failed
<u>Tungsten</u>					
34S	Arc-Plasma	2730	5 min.		Temperature increased
34S	Arc-Plasma	2940	5 min.		Temperature increased
34S	Arc-Plasma	3090	5 min.		Temperature increased
34S	Arc-Plasma	3260	5 min.		Temperature increased
34S	Arc-Plasma	3460	5 min.		Failed
34S	Flame	3450	60 min.	5	Good

TABLE 16  
METALLOGRAPHIC THICKNESS OF BERYLLIDE COATINGS

Coating Treatment Time, Hours	Temp., °F	Alloy	Thickness in mils (0.001")			Remarks
			Coating	Core	Total	
4	1800	Ta	0.7	8.7	0.7	10.1
						Good
4	1900	Ta	1.8	9.2	1.7	12.7
						Satisfactory
4	1900	10% W	1.4	9.6	1.7	12.7
						Satisfactory
1	2000	Ta	1.5			
						Good
2	2000	Ta	2.6	8.9	2.4	13.9
						Good
2	2000	10% W	2.2	8.8	2.1	13.1
						Good
2	2000	Ta	1.4	9.8	1.4	12.6
						Satisfactory
2	2000	10% W	1.7	9.4	1.6	12.7
						Satisfactory
4	2000	Ta	1.2	7.9	2.9	12.0
						Good
4	2000	10% W	2.5	9.3	2.5	14.3
						Satisfactory
4	2000	10% W	1.7	8.9	1.6	12.2
						Satisfactory
8	2000	10% W	2.5	8.4	2.5	13.4
						Satisfactory
4	2000	10% W	4.2	3.2	4.5	11.9
						Oxidized at 2500°F, 93 hrs.

Tantalum uncoated 9.7 mils thick  
Ta-10% W uncoated 10.4 mils thick

TABLE 17

GROWTH OF TANTALUM BERYLLIDES  
Thickness in Mils (0.001")

<u>Zone</u>	<u>As Coated</u>		<u>Vacuum Diffused</u>	<u>2200°F, 25 Hours</u>
	<u>BeS1825</u>	<u>BeS204</u>	<u>BeS1825VD2225</u>	<u>BeS204VD2225</u>
TaBe <sub>12</sub>	1.7	1.4	1.8	2.2
Ta <sub>2</sub> Be <sub>17</sub>	0.24	0.24	0.49	0.74
TaBe <sub>3</sub>	0.12	0.12	2.1	2.5
TaBe <sub>2</sub>	0.74	0.74	0.37	0.24
Ta	8.6	8.7	4.2	4.2
TaBe <sub>2</sub>	0.74	0.74	0.24	0.24
TaBe <sub>3</sub>	0.12	0.12	2.2	2.1
Ta <sub>2</sub> Be <sub>17</sub>	0.24	0.24	0.71	0.74
TaBe <sub>12</sub>	1.6	1.7	1.9	2.7
TOTAL	14.1	14.0	14.01	15.66

additional beryllium was deposited on the samples. The effect of diffusion is obvious. Notable is the rapid growth of  $\text{TaBe}_3$ . This technique was later found to be unnecessary and impractical.

Furnace oxidation results with different coating treatments for the Ta-10 W alloy are given in Table 18. Vapor depositing for 4 hours at 2000°F gave the best oxidation results. Requirements can be met at 2500° and 2600°F and possibly at 2700°F. Experimental difficulties were encountered at 2800°F where reactions were sometimes noted between the  $\text{Al}_2\text{O}_3$  boat and/or platinum. Samples of Ta-10 W were resistant at 2500° to 2800°F for 10 hours and 10 cycles under stresses producing 1 to 15% deformation. Pure tantalum samples stressed during exposure failed in minutes.

The same number of compounds reported to be in the Be-Ta system are found in the layers that comprise the coatings on tantalum, as shown in Figure 19. These layers can be resolved best under polarized light, which makes them optically active. Diffusion effects on core thickness are also illustrated in Figure 19. The coating is primarily  $\text{TaBe}_{12}$  which on exposure to 2500°F or higher with pure tantalum causes rapid diffusion, the substrate receding approximately 0.004" in 15 hours at 2500°F. In Ta-10 W it is somewhat surprising that diffusion occurs much more slowly. After 10 hours at 2800°F, 300 degrees higher, surface recession was only 0.002" after 10 hours. This is in contrast to the aluminide system where little difference in diffusion was found between the two substrate materials. Diffusion in the (Ta-10 W)-beryllide system was somewhat faster than in the aluminide system. It should be noted that the original beryllium concentration gradient is much higher than with aluminum, thus providing more driving force for reaction. Major growth occurred in the  $\text{TaBe}_3$  layer at the expense of  $\text{TaBe}_{12}$ . The  $\text{Ta}_2\text{Be}_{17}$  layer also thickened and the  $\text{TaBe}_2$  layer remained relatively constant. Evidence of twinning was noted in the  $\text{TaBe}_2$ .

No appreciable internal hardening effects were noted after exposure, an increase of 20 KHN occurring in the pure tantalum after 15 hours at 2500°F. The surface compounds are extremely hard, brittle, and difficult to maintain intact during metallographic polishing. All of them are over 1000 KHN and as high as 1900 KHN.

## VII. DISCUSSION

Both the aluminide and beryllide systems have provided very interesting results in protecting pure tantalum and the Ta-10 W

TABLE 18

## FURNACE OXIDATION RESULTS ON BERYLLIDE COATED TANTALUM ALLOY

<u>Temperature,</u> <u>°F</u>	<u>Time,</u> <u>Hours</u>	<u>Cycles</u>	<u>Remarks</u>
<u>Coated 4 hours 1900°F</u>			
1800	4		Failed
1800	23	2	Failed
2200	3		Failed
2200	4		Failed
2500	1		Failed
2500	1		Failed
2500	6	6	Good
2500	7	7	Good
2500	16		Good
2500	21		Good
2600	1		Failed
2600	5	4	Failed
2600	16		Good
2700	16		Good
<u>Coated 2 hours 2000°F</u>			
1800	16		Failed
2200	23	2	Failed
2200	70	7	Failed
2500	2	2	Failed
2500	10	10	Good
2500	16		Good
2500	110		Good
2600	7	6	Failed
2600	9	8	Failed
2600	16		Good
2600	100		Good
2700	16		Good
2700	16		Failed
2800	1		Boat Reaction

TABLE 18 (Cont'd.)

## FURNACE OXIDATION RESULTS ON BERYLLIDE COATED TANTALUM ALLOY

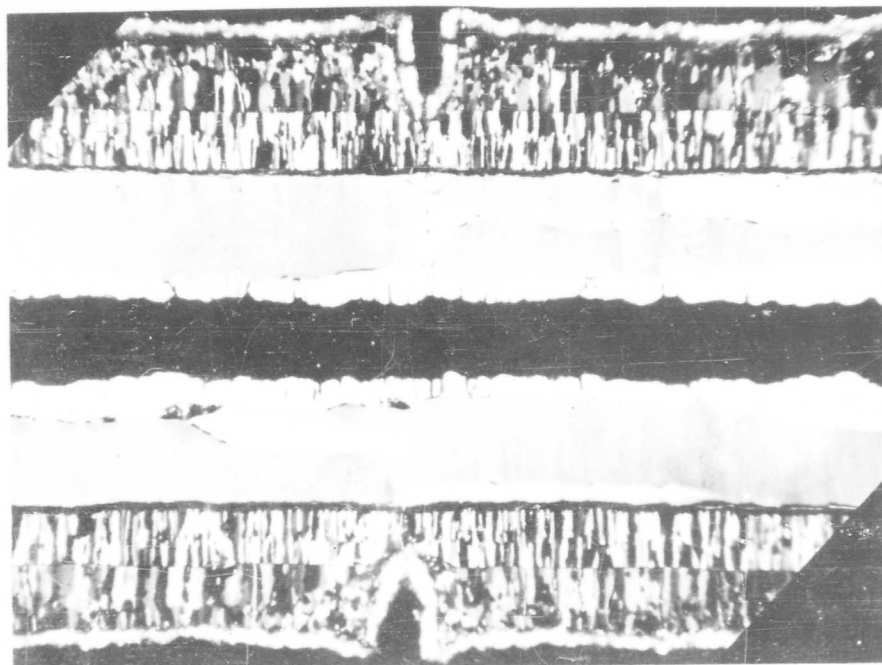
<u>Temperature,</u> <u>°F</u>	<u>Time,</u> <u>Hours</u>	<u>Cycles</u>	<u>Remarks</u>
<u>Coated 4 hours 2000°F</u>			
1800	7	1	Failed
1300	27	3	Failed
2200	7	2	Failed
2200	64		Good
2200	120	13	Failed
2500	10	10	Good
2500	10	10	Good
2500	62	4	Failed
2500	64		Good
2500	93		Good
2600	10	9	Good
2600	10	10	Good
2600	64		Good
2700	7	6	Failed
2700	16		Good
2800	6	5	Boat Reaction
2800	3	2	Boat Reaction
<u>Coated 8 hours 2000°F</u>			
1800	20	1	Failed
2200	64		Good
2200	104	14	Good
2500	10	10	Good
2500	24	23	Failed
2500	64		Good
2600	6	6	Failed
2600	10	10	Good
2600	16		Good
2600	65		Good

TABLE 18 (Cont'd.)

## FURNACE OXIDATION RESULTS ON BERYLLIDE COATED TANTALUM ALLOY

<u>Temperature,</u> <u>°F</u>	<u>Time,</u> <u>Hours</u>	<u>Cycles</u>	<u>Remarks</u>
<u>Coated 4 hours 2500°F</u>			
<u>Ta-10 Hf-5 W Alloy</u>			
2500	6	5	Good
2500	20	5	Failed
2500	64		Good
2700	6	5	Failed
2700	16		Good
2700	<64		Failed

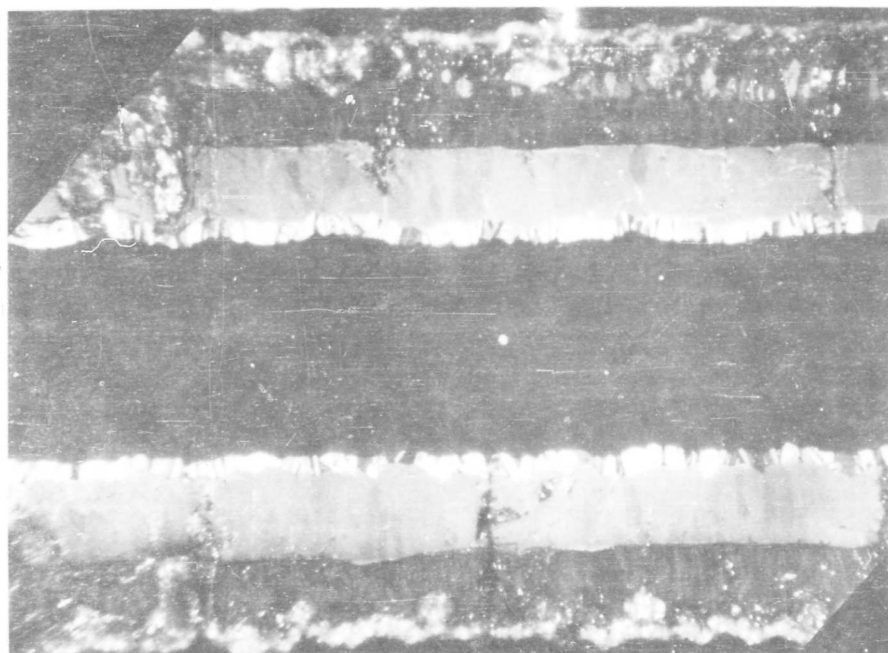




Knoop   Mils

← TaBe <sub>12</sub>	1.6
← Ta <sub>2</sub> Be <sub>17</sub>	1.6
← TaBe <sub>3</sub>	2.9
← TaBe <sub>2</sub>	0.74
← Ta	160   1.9

Plate No. 23664      Sample Be221-1      200X  
Oxidized 10 hours 2500°F      Pure Ta



← Ta <sub>2</sub> Be <sub>17</sub>	2.1
← TaBe <sub>3</sub>	2.4
← TaBe <sub>2</sub>	0.49
← Ta	5.3

Plate No. 23748      Sample TBeS204-4      200X  
Oxidized 10 hours 2800°F      Ta-10 W

Fig. 19. Beryllium coated samples after oxidation for ten hours.

alloy sheet from air oxidation for 10 hours with thermal cycling at temperatures up to 2800°F or higher. Particular emphasis is laid on the Sn-Al system since it is so simple and practical to use and appears to have considerable flexibility of application. It is also very reliable over a broad temperature range and shows evidence of self-healing characteristics and resistance to abuse in handling after coating.

The role of tin is hard to define and considerable investigative work could be done in tracing the migration and concentration of tin-aluminum in and through the several coating layers. It is also difficult to explain the mechanism of repletion of the protective  $\text{Al}_2\text{O}_3$  film and whether or not the oxide is the sole means of protection; it certainly appears to be.

Samples of the Ta-10 W alloy coated with 348 have been investigated by X-ray diffraction techniques. As coated the structure is mainly  $\text{TaAl}_3$ , Al, Sn, and some amounts of  $\text{Ta}_2\text{Al}$ . A sample after being oxidized for 64 hours at 2700°F contained an  $\text{Al}_2\text{O}_3$  surface film. Grinding off successive layers revealed mostly  $\text{Ta}_2\text{Al}$  plus  $\text{Al}_2\text{O}_3$  and beta tin. These structures corroborate the phases seen in the microstructure.

A suggested mechanism for oxidation protection of tin-aluminum coated tantalum could be the following. An absolute necessity is the presence of  $\text{TaAl}_3$  during initial exposure to oxidizing conditions. Many samples of Ta, Ta-10 W and Ta-10 Hf-5 W during or after coating have been heated to 2000°F or higher under non-oxidizing conditions. In every case oxidation failure is immediate. In the later case before oxidation all or most of the  $\text{TaAl}_3$  has been converted to  $\text{Ta}_2\text{Al}$ . This compound has no oxidation resistance. These data prove the first premise. After the  $\text{TaAl}_3$  has formed an  $\text{Al}_2\text{O}_3$  oxide layer and been converted to  $\text{Ta}_2\text{Al}$  by diffusion during oxidation, enough Al (less than 5%) is dissolved in the tin from the  $\text{Ta}_2\text{Al}$  to react with oxygen at the tin- $\text{Al}_2\text{O}_3$  interface or tin-air interface, causing growth of the  $\text{Al}_2\text{O}_3$  layer or to heal any defect in the oxide. There is no indication of the  $\text{Al}_2\text{O}_3$  being formed at the  $\text{Ta}_2\text{Al}$  interface and floating through the tin to the oxide layer. The small solubility of Al in the Sn must confer the self-healing characteristics. Tin-free aluminum compositions such as G-14 do not have this ability. Whenever the  $\text{Al}_2\text{O}_3$  layer is broken by thermal shock, etc., the oxygen can react with  $\text{Ta}_2\text{Al}$  and cause immediate failure without healing the oxide. This theory seems to explain all observed oxidation phenomena with pure Ta and the Ta-10 W alloy. A similar explanation can be used for the

ability of Sn-Al coating to protect certain columbium alloys. Although  $\text{CbAl}_3$  is much more thermally stable than  $\text{TaAl}_3$ , it does have a finite life at 2500°F.

A harmful unknown phase is present in the Sn-Al coated Ta-10 Hf-5 W alloy. Therefore, it may be preferable to use a Navy-Westinghouse developed Ta alloy, Ta-8 W-2 Hf, since it has equal high temperature properties and preliminary results at Sylcor have shown it can be coated successfully. This is probably due to the lower hafnium content.

It is not suggested that the 50 Sn-50 Al coating composition is the best for tantalum alloys. Chemical analysis of the 348 coating after diffusion shows that 34% Al is still present in the Sn-Al phase. The Al can still react with Ta to form Ta-Al compounds. Good results were obtained with initial tests of the 408 composition (75 Sn-25 Al) which has less aluminum. Core regression should be less than with the 348 coating. Possibly even better would be: (1) a first coat of Sn-25 Al, (2) diffuse, and (3) a second coat of Sn-10 Al and diffuse. The variations are almost infinite. A duplex coating technique seems to be required. The reason is not known but may be associated with reliability of more complete coverage.

The tantalum-beryllium system also merits more work, since BeO has a higher melting point than  $\text{Al}_2\text{O}_3$ . A diluent should be found for the beryllides similar to tin with the aluminides. Although diffusion effects are more rapid with beryllium, tungsten alloying appears to slow down the process and give hope that additions higher than 10% would be even more effective. It is recognized that elevated temperature oxidation rates of the beryllides is adversely affected by moisture<sup>(5)</sup>. However, the refractory nature of BeO and the possible gains in protectiveness make it attractive for further study along the lines suggested.

#### VIII. SUMMARY

The objectives of this program were met and surpassed. Aluminides were produced on tantalum materials that would protect the core from oxidation for a minimum of 10 hours with 10 thermal cycles from test temperature to ambient under a stress producing at least 2% elongation without failure. A dipped and diffused coating of Al-5 Ti-5 Cr (G14) would give 10 hours with 10 cycles protection in the range 2500° to 2700°F to ambient. Lower temperature properties

were relatively poor. A 90 Sn-10 Al (G21) dip coating would meet these same requirements up to 2600°F without the poor low temperature phenomenon. The duplex spray coating of 50 Sn-50 Al (34S) would protect tantalum and the Ta-10 W alloy up to 100 hours with 100 cycles to 2800°F. No anomalous behavior was found between -320°F to 3000°F or possibly 3400°F with this coating. Diffusion appears to be a problem with Ta-10 Hf-5 W. The 34S coating on the Ta-10 W alloy can withstand high velocity hot air streams for reasonable times.

Feasibility of 34S protection was shown for a columbium alloy, molybdenum, and tungsten.

Good protection was obtained with beryllide coatings on Ta-10 W in the range of 2500° to 2700°F. Samples behaved poorly in the range 1800° to 2200°F. Brittleness of the beryllides seems to be the main problem with this type coating.

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<p>SYLOR DIVISION, SYLVANIA ELECTRIC PRODUCTS, INC., Bayside, New York. HIGH TEMPERATURE OXIDATION RESISTANT COATINGS FOR TANTALUM BASE ALLOYS. BY Dean D. Lawthers and L. S. Sams, October 1961, 73p. incl. figs., tables and refs. (Project 731; Task 73512) (ASD TR 61-233) (Contract AF 33(616)-7462)</p> <p>Unclassified Report</p> <p>Aluminide and beryllide coatings were investigated for pure tantalum, a commercial Ta-10 W alloy, and a ternary alloy under development. Coatings were applied by dipping, packing, cold spraying, and vapor deposition. Oxidation resistance was evaluated by furnace testing, resistance heating, and flame testing. Internal hardening and diffusion effects were also studied. A Sn-Al coating</p> <p>( over )</p>	<p>UNCLASSIFIED</p>
<p>was developed, which has excellent oxidation resistance for 10 hours to at least 3000°F. Evaluation was successfully extended to arc-plasma tests which also showed the feasibility of protecting molybdenum and tungsten. A similar but minor effort investigation was performed with a columbium alloy which could be protected to 500°F for at least 10 hours.</p>	<p>UNCLASSIFIED</p>
<p>SYLOR DIVISION, SYLVANIA ELECTRIC PRODUCTS, INC., Bayside, New York. HIGH TEMPERATURE OXIDATION RESISTANT COATINGS FOR TANTALUM BASE ALLOYS. BY Dean D. Lawthers and L. S. Sams, October 1961, 73p. incl. figs., tables and refs. (Project 731; Task 73512) (ASD TR 61-233) (Contract AF 33(616)-7462)</p> <p>Unclassified Report</p> <p>Aluminide and beryllide coatings were investigated for pure tantalum, a commercial Ta-10 W alloy, and a ternary alloy under development. Coatings were applied by dipping, packing, cold spraying, and vapor deposition. Oxidation resistance was evaluated by furnace testing, resistance heating, and flame testing. Internal hardening and diffusion effects were also studied. A Sn-Al coating</p> <p>( over )</p>	<p>UNCLASSIFIED</p>
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Unclassified Report

Aluminide and beryllide coatings were investigated for pure tantalum, a commercial Ta-10 W alloy, and a ternary alloy under development. Coatings were applied by dipping, packing, cold spraying, and vapor deposition. Oxidation resistance was evaluated by furnace testing, resistance heating, and flame testing. Internal hardening and diffusion effects were also studied. A Sn-Al coating

( over )

was developed, which has excellent oxidation resistance for 10 hours to at least 3000°F. Evaluation was successfully extended to arc-plasma tests which also showed the feasibility of protecting molybdenum and tungsten. A similar but minor effort investigation was performed with a columbium alloy which could be protected to 2500°F for at least 10 hours.

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SYLCON DIVISION, SYLVANIA ELECTRIC PRODUCTS, INC., Bayside, New York. HIGH TEMPERATURE OXIDATION RESISTANT COATINGS FOR TANTALUM BASE ALLOYS. BY Dean D. Lewthers and L. S. Sams. October 1961. 73p. Incl. figs., tables and refs. (Project 751; Task 73512) (ASD TH 61-233) (Contract AF 33(616)-7462)

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<p>SYLOR DIVISION, SYLVANIA ELECTRIC PRODUCTS, INC., Bayside, New York. HIGH TEMPERATURE OXIDATION RESISTANT COATINGS FOR TANTALUM BASE ALLOYS. BY Dean D. Lavthers and L. Sams, October 1961, 73p. incl. figs., tables and refs. (Project 731; Task 73512) (ASD TR 61-233) (Contract AF 33(616)-7462)</p> <p>Unclassified Report</p> <p>Aluminide and beryllide coatings were investigated for pure tantalum, a commercial Ta-10 W alloy, and a ternary alloy under development. Coatings were applied by dipping, packing, cold spraying, and vapor deposition. Oxidation resistance was evaluated by furnace testing, resistance heating, and flame testing. Internal hardening and diffusion effects were also studied. A Sn-Al coating</p> <p>( over )</p>	<p>UNCLASSIFIED</p>
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<p>UNCLASSIFIED</p>	<p>was developed, which has excellent oxidation resistance for 10 hours to at least 3000°F. Evaluation was successfully extended to arc-plasma tests which also showed the feasibility of protecting molybdenum and tungsten. A smaller but minor effort investigation was performed with a columbium alloy which could be protected to 500°F for at least 10 hours.</p> <p>UNCLASSIFIED</p>

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<p>was developed, which has excellent oxidation resistance for 10 hours to at least 3000°F. Evaluation was successfully extended to arc-plasma tests which also showed the feasibility of protecting molybdenum and tungsten. A similar but minor effort investigation was performed with a columbium alloy which could be protected to 2500°F for at least 10 hours.</p>	<p>UNCLASSIFIED</p>



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